

09/029977

029, 408

Attorney's Docket No. : GK-GEY-1020A  
U.S. Application No. :  
International Application No. : PCT/EP96/04016  
International Filing Date : SEPTEMBER 12, 1996 (12 SEPTEMBER 1996)  
Priority Date Claimed : SEPTEMBER 12, 1995 (12 SEPTEMBER 1995)  
Title of Invention : PROCESS FOR PRODUCING MOULDINGS WITH A BARRIER  
LAYER MADE OF BIODEGRADABLE MATERIAL, AND  
MOULDINGS PRODUCED ACCORDING TO THIS PROCESS  
Applicant(s) for (DO/EO/US) : Andreas Gerhard BAAR, Wolfgang GEBEL, Ute IMHOF and  
Hannelore MIHALIK

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. This express request to begin national examination procedures [35 U.S.C. 371 (f)] at any time rather than delay examination until the expiration of the applicable time limit set forth in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. A copy of the International Application as filed [35 U.S.C. 371(c)(2)]
  - a)  is transmitted herewith (required only if not transmitted by the International Bureau)
  - b)  has been transmitted by the International Bureau
  - c)  is not required, as the application was filed in the United States Receiving Office (RO/US)
6. A translation of WO 97/10293 (~~the International Application~~) into English [35 U.S.C. 371(c)(2)]
7. Amendments to the claims of the International Application under PCT Article 19 [35 U.S.C. 371(c)(3)]
  - a)  are transmitted herewith (required only if not transmitted by the International Bureau)
  - b)  have been transmitted by the International Bureau
  - c)  have not been made; however, the time limit for making such amendments has **NOT** expired.
  - d)  have not been made and will not be made
8. A translation of the amendments to the claims under PCT Article 19 [35 U.S.C. 371(c)(3)]
9. An oath or declaration of the inventor(s) [35 U.S.C. 371(c)(4)] **TO FOLLOW**
10. A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 [35 U.S.C. 371(c)(5)]

**Items 11. to 16. below concern other document(s) or information included:**

11. An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98
12. An Assignment document for recording. A separate cover sheet (PTO-1595) in compliance with 37 CFR 3.28 and 3.31 is included.
13. **A FIRST** preliminary amendment and Abstract  
 **A SECOND** or **SUBSEQUENT** preliminary amendment
14. A substitute specification
15. A change of power of attorney and/or address letter
16. (other items or information)

EXPRESS MAIL No.: **EM 165 841 815 US** Deposited: **March 12, 1998**

I hereby certify that this correspondence is being deposited with the United States Postal Service Express mail under 37 CFR 1.10 on the date indicated above and is addressed to: BOX PCT, ASSISTANT Commissioner for Patents, Washington, DC 20231.

*Ruth Montalvo*

/Ruth Montalvo Date: **March 12, 1998**

17.  The following fees are submitted:

BASIC NATIONAL FEE [37 CFR 1.492(a)(1)-(5)]:

CALCULATIONS PTO USE ONLY

<input checked="" type="checkbox"/> Search Report has been prepared by the EPO or JPO.....	\$ 930.00		
<input type="checkbox"/> International preliminary examination fee paid to USPTO [37 CFR 1.482].....	\$ 720.00		
<input type="checkbox"/> No International preliminary examination fee paid to USPTO [37 CFR 1.482] but International search fee paid to USPTO [37 CFR 1.445(a)(2)].....	\$ 790.00		
<input type="checkbox"/> Neither International preliminary examination fee [37 CFR 1.482] nor International search fee [37 CFR 1.445(a)(2)] paid to USPTO.....	\$ 1,070.00		
<input type="checkbox"/> International preliminary examination fee paid to USPTO [37 CFR 1.482] and all claims satisfied provisions of PCT Article 33(1)-(4).....	\$ 98.00		

ENTER APPROPRIATE BASIC FEE AMOUNT:

\$ 930.00

Claims	Number filed	Number extra	Rate	
Total Claims (Prel.Amt)	59 -20=	39	x \$ 22. =	\$ 858.00
Indep. Claims	4 -03=	1	x \$ 82. =	4 82.00
Multiple Dependent Claim(s) (if applicable)		+	\$ 270. =	

TOTAL OF ABOVE CALCULATIONS:

\$ 1,870.00

Surcharge of \$130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months  
from the earliest claimed priority date [37 CFR 1.492(e)]

TOTAL OF ABOVE CALCULATIONS:

\$ 1,870.00

Reduction by ½ for filing by small entity, if applicable. Verified Small Entity Statement must  
be filed. [Note 37 CFR 1.9, 1.27, 1.28]

SUBTOTAL: \$ 1,870.00

Processing fee of \$130.00 for furnishing the English Translation later than [ ] 20  
[ ] 30 months from the earliest claimed priority date [37 CFR 1.492(f)]

TOTAL NATIONAL FEE: \$ 1,870.00

Fee for recording the enclosed assignment [37 CFR 1.21(h)] The assignment must be  
accompanied by an appropriate cover sheet (PTO-1595) [37 CFR 3.28, 3.31]. \$ 40.00 per property +

TOTAL FEES TO BE CHARGED: \$ 1,870.00

AMOUNTS TO BE  
REFUNDED OR CHARGEDREFUNDED  
CHARGED \$ Enclosed is a check in the amount of \$ 1,870.00. Please charge my Deposit Account No. 13-0025 in the amount of \$ .00 to cover the above fees. The Commissioner is hereby authorized to charge the deposit account the above-mentioned fees and any other fees required  
with this submission or to credit any overpayment to Deposit Account No: 13-0025. A duplicate of this form is enclosed.NOTE: Where an appropriate time limit under 36 CFR 1.494 or 1.495 has not been met, a petition to revive [37 CFR 1.137(a) or (b)]  
must be filed and granted to restore the application to pending status.

## SEND ALL CORRESPONDENCE TO:

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25.116

March 12, 1998

Reg. No.

Date

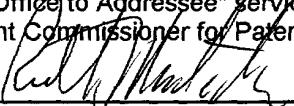
105 Rec'd PCT/PTO 12 MAR 1998

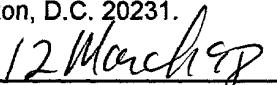
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Ruth Montalvo

  
Date

Docket No.: GK-GEY-1020A

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant : Andreas Gerhard BAAR, Wolfgang GEBEL,  
Ute IMHOF and Hannelore MIHALIK

Serial No. : Unknown (Int'l Appln. PCT/EP96/04016  
filed September 12, 1996)

Filed : Concurrently herewith

For : PROCESS FOR PRODUCING MOULDINGS WITH  
A BARRIER LAYER MADE OF BIODEGRADABLE  
MATERIAL, AND MOULDINGS PRODUCED ACCORDING  
TO THIS PROCESS

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Please amend the above-identified application, filed simultaneously herewith,  
as follows:

IN THE TITLE:

Please correct the spelling of "MOULDING" to --MOLDINGS--.

IN THE SPECIFICATION:

Page 1, line 3, after this line insert the headings:

--BACKGROUND OF THE INVENTION--;

--a) Field of the Invention--;

line 10, after this line insert the heading:

--b) Description of the Related Art--;

Page 3, line 24, after this line insert the heading:

--OBJECT AND SUMMARY OF THE INVENTION--;

line 25, after "the" (first occurrence) insert --primary--;

Page 7, line 26, change "the" to --with--;

Page 8, line 23, after this line insert the heading:

--BRIEF DESCRIPTION OF THE DRAWINGS--;

after this line insert the paragraph:

--In the drawings:--;

Page 10, line 11, after "softeners;" insert --and--;

line 14, after this line insert the heading:

--DESCRIPTION OF THE PREFERRED EMBODIMENTS--;

Page 20, line 29, change "The" to --As--;

Page 33, last line, after this line insert the following paragraph:

--While the foregoing description and drawings represent the present invention, it will be obvious to those skilled in the art that various changes may be made therein without departing from the true spirit and scope of the present invention.--

**IN THE CLAIMS:**

Preceding "1." change "Patent Claims" to --What is claimed is:--.

Amend the claims as follows:

1. (amended). [Process] In a process for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material, water, and starch and is baked in a baking mold so as to form a fiber material-starch composite, [characterized in that] an improvement comprising the steps of: using a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm [is used] as fiber material[,] ; and producing the molded body [is produced] by impregnation with a biodegradable, hydrophobic boundary layer, wherein the boundary layer is formed from cellulose acetate and/or cellulose acetate propionate without softeners.

2. (Amended) [Process] In a process for producing molded bodies with a

barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material and starch and is baked in a baking mold so as to form a fiber material-starch composite, [characterized in that] an improvement comprising the steps of:

using a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm [is used] as fiber material; and producing the molded body [is produced] by means of a foil coating formed by applying a foil based on polyester, polyester amide or polylactic acid to the baked molded body so as to form a biodegradable, liquid-tight boundary layer.

3. (Amended) [Process] The process according to claim 2, [characterized in that] including the step of forming the biodegradable boundary layer [is formed] without softeners.

Claim 4, line 1, change "Process" to --The process--; delete "or 3"; change "characterized in that" to --wherein--.

Claim 5, line 1, change "Process" to --The process--; change "characterized in that" to --wherein--.

6. (Amended) [Process] The process according to [at least one of the preceding claims 2 to 4] claim 2, [characterized in that] wherein the foil coating is

carried out by vacuum heat sealing or compression heat sealing and/or with heat treatment.

7. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 6] claim 1, [characterized in that] wherein the impregnation or foil coating is elastic and is [preferably] applied directly to the molded body without adhesion promoters.

8. (Amended) [Process] The process according to [at least one of the preceding claims 2 to 4, 6 or 7] claim 2, [characterized in that] wherein the foil is heated at its underside intended for adhesive connection with the molded body.

9. (Amended) [Process] The process according to [at least one of the preceding claims 2 to 4, 6 to 8] claim 2, [characterized in that] wherein the foil is prestretched prior to application to the molded body by means of a male die, especially for hollow molded bodies.

10. (Amended) [Process] The process according to [at least one of the preceding claims 2 to 4, 6 to 9] claim 2, [characterized in that] wherein at least the adhesion surface of the molded body to be coated with foil is preheated prior to coating with foil to the melting temperature of the foil and the foil is then applied thereto.

11. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 10] claim 1, [characterized in that] wherein the hydrophobic boundary layer is formed by impregnation or a liquid-tight barrier layer by applying a foil to the molded body when it is still hot from the preceding baking process and the molded body is conditioned simultaneously or subsequently.

Claim 12, line 1, change "Process" to --The process--; change "at least one of the preceding claims 1 to 11" to --claim 1--.

13. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 12] claim 1, [characterized in that] wherein the molded body is provided with the boundary layer on the inside and/or on the outside.

Claim 14, line 1, change "Process" to --The process--; change "claims 1 to 5, 7, or 11 to 13" to --claim 1--; change "characterized in that" to --wherein--.

Claim 15, line 1, change "Process" to --The process--; change "at least one of the preceding claims 1 to 14" to --claim 1--.

Claim 16, line 1, change "Process" to --The process--; change "15" to --1--; change "characterized in that" to --wherein--.

17. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 16] claim 1, [characterized in that] wherein an inner boundary layer of the molded body has a greater resistance to moisture and/or temperature and/or fats and/or is more tightly sealed against gas and aromas than an outer surface of the molded body, especially an outer surface provided with a hydrophobic boundary layer.

18. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 17] claim 1, [characterized in that] wherein the molded body is provided along an inner surface with a hydrophobic impregnation or coating which is thicker than that on its outer surface.

19. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 18] claim 1, [characterized in that] wherein the molded body is baked in the baking mold, subsequently removed from the baking mold and introduced into a further device within the baking apparatus for applying the hydrophobic impregnation or foil coating.

20. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 19] claim 1, [characterized in that] wherein for subsequent impregnation or coating of the molded body, use is made of process heat of the baking

mold, preferably within the baking mold, or residual heat of the molded article from the preceding baking process to heat the molded body and/or to heat a hydrophobic medium, especially a coating foil, to be applied to the molded body.

21. (Amended) [Process] The process according to [at least one of the preceding claims 1, 5, 7 or 11 to 20] claim 1, [characterized in that] wherein a solution containing cellulose acetate and/or cellulose acetate propionate as hydrophobic substance and a solvent, especially a volatile solvent, is used for impregnation.

22. (Amended) [Process] The process according to [at least one of the preceding claims 2 to 4, 6 to 10] claim 2, [characterized in that] wherein the foil is trimmed at the coated molded body after the molded body is removed from the baking mold in conjunction with a severing of overlapping material resulting from the steam discharge ducts of the baking mold.

23. (Amended) [Process] The process according to [at least one of claims 1 to 23] claim 1, [characterized in that] wherein fiber-containing raw material which is pulped or defibrated in particular by prior comminution is used to form the fiber material.

24. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 23] claim 1, [characterized in that] wherein the fiber-containing

raw material is waste paper, recycled material, especially deinked waste paper, biodegradable fiber material such as production waste containing cellulose fibers, especially wood pulp or paper pulp, beet chips or the like.

25. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 24] claim 1, [characterized in that] wherein the fiber material is formed directly from biodegradable fibers or fiber bundles.

Claim 26, line 1, change "Process" to --The process--; change  
"characterized in that" to --wherein--.

27. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 25] claim 1, [characterized in that] wherein the starch used is native starch and/or pregelatinized starch or modified starch.

Claim 28, line 1, change "Process" to --The process--; change  
"at least one of the preceding claims 1 to 27" to --claim 1--.

29. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 28] claim 1, [characterized in that] wherein water is added in a ratio of approximately 8 : 1 [, preferably 2.5:1,] in relation to the dry mass of the fiber-containing raw material, especially waste paper, for forming the viscous mass.

30. (Amended) [Process] The process according to [at least one of the preceding claims 27 to 29] claim 27, [characterized in that] wherein the proportion of pregelatinized starch or modified starch of the total starch in the viscous mass is approximately 20 to 75 percent by weight.

31. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 30] claim 1, [characterized in that] wherein the proportion of starch to water in the viscous mass is approximately 1:10 to 1:1[, preferably 1:3 to 1:2], and water is subsequently added in the form of modified starch or starch which is pregelatinized accompanied by excess water to form the viscous mass.

32. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 31] claim 1, [characterized in that] wherein the portion of fiber material in the viscous mass is 10 percent by weight to 30 percent by weight, the proportion of starch in the viscous mass is 5 percent by weight to 40 percent by weight, and the proportion of water in the viscous mass is 70 percent by weight to 40 percent by weight.

33. (Amended) [Process] The process according to [at least one of the preceding claims 27 to 32] claim 27, [characterized in that] wherein the proportion of pregelatinized starch or modified starch in the viscous mass is 1 percent by weight to 13 percent by weight.

34. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 33] claim 1, [characterized in that] wherein the fiber-containing raw material, especially waste paper, is comminuted and subsequently broken down to its fiber structure in the presence of water and with the addition of starch, [preferably native starch,] and the moldable viscous mass is formed and subsequently baked to form molded bodies.

35. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 33] claim 1, [characterized in that] wherein the fiber-containing raw material, especially waste paper, is comminuted in the presence of water and is subsequently broken down to its fiber structure with the addition of starch [, preferably native starch,] and the moldable viscous mass is formed and subsequently baked to form molded bodies.

36. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 35] claim 1, [characterized in that] wherein the viscous mass is metered prior to baking.

37. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 36] claim 1, [characterized in that] wherein the fiber-containing raw material or the fiber material that is used directly is analyzed with respect to its fiber length and starch proportion and is graded prior to or after the comminuting

process.

38. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 37] claim 1, [characterized in that] wherein the native starch is added in partially already during a comminution of the fiber-containing raw material, especially as a premix with filler materials.

39. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 40] claim 1, [characterized in that] wherein the starch is at least partially added to the fiber-containing raw material[, at least partially,] during the comminuting process and/or during a subsequent drying process or wet mixing process and/or during a subsequent equalizing mixing and kneading process as native starch and/or modified starch and/or pregelatinized starch.

40. (Amended) [Process] The process according to [at least one of the preceding claims 27 to 39] claim 27, [characterized in that] wherein the starch is introduced as pregelatinized starch and as native starch and at least a filler material is added into the native starch prior to adding in the comminuting process of the fiber-containing raw material or in the subsequent equalizing mixing and kneading process.

41. (Amended) [Process] The process according to [at least one of the preceding claims 27 to 40] claim 27 , [characterized in that] wherein the pregelatinized

starch is added directly to the mixing and kneading process for forming the viscous mass.

42. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 41] claim 1, [characterized in that] wherein the formation of the viscous mass is carried out using fiber-containing raw material of different grades of comminution and different fiber lengths.

43. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 42] claim 1, [characterized in that] wherein the mass is heated before introducing it into the baking mold.

44. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 43] claim 1, [characterized in that] wherein the at least two baking plates of the baking mold are locked relative to one another at the beginning of the baking process while ensuring a constant distance between the upper baking plate and lower baking plate during the baking process, wherein the distance between the inner surfaces of the baking plates which face one another and between which the mass is located determines the wall thickness of the baked molded body.

45. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 44] claim 1, [characterized in that] wherein a controlled discharge

of steam from the baking mold is carried out during the baking process.

Claim 46, line 1, change "Process" to --The process--; change  
"characterized in that" to --wherein--.

47. (Amended) [Process] The process according to [at least one of the preceding claims 1 to 46] claim 1, [characterized in that] wherein long fibers or long fiber bundles with a length of up to 50 mm are [preferably] used for thick-walled and/or large-area molded bodies, especially fibers or fiber bundles with a fiber length or fiber bundle length in the range of 10 mm to 50 mm in a mixture with short fibers or short fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 20 mm.

48. (Amended) [Process] [Molded] A molded body, especially a molded body for packaging, made from biodegradable material, comprising:

biodegradable fiber material, starch and a residual proportion of water, especially produced in accordance with [the] a process [according to at least one of the preceding claims 1 to 47] for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material, water, and starch and is baked in a baking mold so as to form a fiber material-starch composite, said process comprising the steps of using a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm as fiber material and producing the

molded body by impregnation with a biodegradable, hydrophobic boundary layer,  
wherein the boundary layer is formed from cellulose acetate and/or cellulose acetate  
propionate without softeners; [, characterized in that the]

said molded body [has] having:

a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 50 mm, and [has] having, on at least one side, a biodegradable hydrophobic boundary layer which is formed based on cellulose acetate or cellulose acetate propionate without softeners or has a coating foil based on polyester, polyester amide or polylactic acid.

Claim 49, line 1, change "Molded" to --The molded--; change  
"characterized in that" to --wherein--.

Claim 50, line 1, change "Molded" to --The molded--; change  
"characterized in that" to --wherein--;  
line 2, delete "preferably".

51. (Amended) [Molded] The molded body according to [at least one of the preceding claims 48 to 50] claim 48, [characterized in that] wherein the proportion of fiber material to starch is 4:1 to 1:4.

52. (Amended) [Molded] The molded body according to [at least one of the

preceding claims 48 to 51] claim 48, [characterized in that] wherein the molded body contains a proportion of pregelatinized starch or modified starch.

53. (Amended) [Molded] The molded body according to [at least one of the preceding claims 48 to 51] claim 48, [characterized in that] wherein the molded body contains a mixture of, in particular, long, loosened fiber bundles and, in particular, short individual fibers or fiber bundles.

Add new claims 54-59 reading as follows:

--54. A molded body, especially a molded body for packaging, made from biodegradable material, comprising:  
biodegradable fiber material, starch and a residual proportion of water, especially produced in accordance with a process for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material and starch and is baked in a baking mold so as to form a fiber material-starch composite, said process comprising the steps of using a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm as fiber material and producing the molded body by means of a foil coating formed by applying a foil based on polyester, polyester amide or polylactic acid to the baked molded body so as to form a biodegradable, liquid-tight boundary layer;

said molded body having:

a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 50 mm, and having on at least one side, a biodegradable hydrophobic boundary layer which is formed based on cellulose acetate or cellulose acetate propionate without softeners or has a coating foil based on polyester, polyester amide or polylactic acid.

55. The molded body according to claim 54, wherein the molded body is covered on all sides.

56. The molded body according to claim 55, wherein long fibers or long fiber bundles with a length of up to 50 mm are used for thick-walled and/or large-area molded bodies, especially fibers or fiber bundles with a fiber length or fiber bundle length in the range of 10 mm to 50 mm in a mixture with short fibers or short fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 20 mm.

57. The molded body according to claim 54, wherein the proportion of fiber material to starch is 4:1 to 1:4.

58. The molded body according to claim 54, wherein the molded body contains a proportion of pregelatinized starch or modified starch.

59. The molded body according to claim 54, wherein the molded body contains a mixture of, in particular, long, loosened fiber bundles and, in particular, short individual fibers or fiber bundles.--

IN THE ABSTRACT OF THE DISCLOSURE

Please add into the application the enclosed new Abstract of the Disclosure.

REMARKS

Claims 1-53 have been amended to improve their form and to eliminate multiple dependencies in order to reduce the filing fee. Claims 54-59 have been added.

The specification has also been amended and a new abstract of the disclosure has been added to conform to U.S. format.

An early and favorable action on the merits is respectfully requested.

Respectfully submitted,

By: Gerald H. Kiel  
Gerald H. Kiel  
Reg. No. 25,116

Dated: March 12, 1998

McAULAY FISHER NISSEN  
GOLDBERG & KIEL, LLP  
261 Madison Avenue  
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Tel. (212) 986-4090  
GHK:jl

Enc.: Abstract of the Disclosure

## ABSTRACT OF THE DISCLOSURE

A process for the production of molded bodies from completely decomposable, biodegradable material with a barrier layer, especially for packaging is disclosed. The biodegradable material uses a viscous mass which contains biodegradable fiber material, water, and starch and is baked in a baking mold so as to form a fiber material-starch composite. A mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm as fiber material. The molded body is produced by impregnation with a biodegradable, hydrophobic boundary layer. The boundary layer is formed from cellulose acetate and/or cellulose acetate propionate without softeners. A molded body produced from the process is also disclosed.

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PROCESS FOR PRODUCING MOULDINGS WITH A BARRIER LAYER MADE OF  
BIODEGRADABLE MATERIAL, AND MOULDINGS PRODUCED ACCORDING TO  
THIS PROCESS

The invention is directed to a process for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material and to a molded body according to this process. The invention is directed particularly to a process for the production of molded bodies for use as recyclable, biodegradable packaging which contain biodegradable fiber material and which are sufficiently resistant, for example, to hold moist foods and which correspond to climatic requirements in the fields of use.

Large quantities of used paper as well as cellulose fiber-containing and starch-containing production waste are produced in homes and in industry. For reasons of environmental protection and preservation of natural resources, recycled waste paper is also reused to an increasing extent as starting material for paper production.

On the other hand, there is an urgent need, especially in the packaging field but also in other fields, to use molded bodies which do not place more than a small burden on the waste industry, are recyclable and biodegradable substantially without residue, e.g., within the framework of composting. Waste paper and cellulose fiber-containing and starch-containing production waste, e.g., mechanical wood pulp or paper pulp as well as deinked waste paper (deinking material), are therefore taken into consideration as basic materials for the production of molded bodies as packaging. In the packaging of foodstuffs, products produced from recycled waste paper are subjected to a special surface treatment in order to meet hygienic requirements. When untreated waste paper issuing from industrial processing is used exclusively, a treatment of the type mentioned above can be dispensed with while conforming to legally prescribed production conditions.

For example, molded packaging is produced by the fiber casting process which is based on the fundamentals of conventional paper production. For this purpose, fiber suspensions are prepared from comminuted waste paper and water and are delivered to mold sieves. The water is subsequently drawn off and the molded article is dried; it may also be subjected to a compacting pressing treatment.

A disadvantage herein consists in that it is very complicated to carry out this process such as it is known, for example, from DE 40 35 887 and the environment is burdened due to the high water requirement. Also, there have recently been a substantial number of attempts to replace molded packaging manufactured from plastic, in particular molded packaging suitable for packaging of foodstuffs, by products based on waste paper.

For example, DE-OS 39 23 497 discloses a multiple-layer packing shell for foodstuff having a carrier layer that is predominantly composed of recycled material based on waste paper. A proportion of foamed plastic is needed as a binder for this purpose. The production of such packaging shells requires a relatively complicated, multiple-step process in which comminuted waste paper material is fed via an extruder to a web-like carrier layer and is subsequently molded in composite action with a cover layer by deep drawing or pressing to form packaging shells. During production, it is necessary to carry out a controlled and metered addition of plastics granules as binder in the carrier layer. It is disadvantageous that, because of the use of plastic, a packaging shell of this kind is not comprised entirely of decomposable organic material. Moreover, production is cost-intensive due to the complicated process management.

Packaging based on waffle dough as edible packaging or as inedible packaging, depending on the added materials, is also known (EP 513 106). In this regard, difficulties arise with respect to the longevity, elasticity, resistance to breaking, and preservability of such packaging.

Finally, it is also known to produce molded packaging material based essentially on starch using modified starch which is melted and subsequently extruded and cooled (EP 0 304 401 B1). However, the product characteristics of the packaging material obtained in this way are not entirely satisfactory for many purposes because of the relatively low resistance to breakage of packaging based on starch and due to its hygroscopic characteristics.

The production of biodegradable packaging based on cellulose and starch and water using the same technology employed in waffle baking is known from WO 95/20628.

When foodstuff is packaged in biodegradable packaging, the barrier effect of such packaging relative to water, steam, grease or fats, oxygen or acidic foodstuffs and aromas plays a significant role. To an overwhelming degree, resistance of biodegradable packaging to such media is only insufficient, so that, in many areas, it is not as useful as disposable packaging made from plastics or plastic composites.

For this reason, it is known for the packaging of liquid foodstuffs to provide a biodegradable packaging material which has an oxygen-blocking core layer of polyvinyl alcohol which is coated on both sides with a biodegradable plastic, e.g., polyhydroxyl butyrate, wherein these biodegradable plastic layers are in turn coated with long-term water-repellant layers, e.g., made from cellulose derivative and/or paper, for protecting against environmental conditions (EP 0 603 876 A1). The production of such composite foils is relatively complicated and does not result in three-dimensional molded bodies.

It is known from EP-A-0 474 095 to provide biodegradable packaging materials with a water-repellant impregnation. WO 95/13734 discloses a process for water-repellant and water-tight coatings of molded bodies based on starch with a film-forming material, e.g., cellulose acetate or cellulose acetate propionate, in which hydrophobic decomposable softeners are used with the hydrophobic film-forming material.

However, it has been shown that for many applications a barrier layer which is completely water-resistant over a longer period of time is not required and may possibly also hinder biodegradability. Further, softeners tend to migrate out of the polymer composite into the foodstuff coming into contact therewith and to reduce resistance to boiling water.

Therefore, the object of the invention is to provide a process for the production of molded bodies with a barrier layer made from biodegradable material which can be carried out economically and simply and to provide a molded body produced in this way, especially for packaging purposes, made from completely decomposable, biodegradable material which is characterized by a high surface quality and low porosity, high structural strength and elasticity, and by sufficient

characteristics of its surface such that it is suitable for holding water-containing and/or grease-containing foodstuffs of different temperatures.

This object is met, according to the invention, in a process of the type mentioned above in that a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm is used as fiber material and the molded body is produced by impregnation with a biodegradable, hydrophobic boundary layer, wherein the boundary layer is formed from a plastics material without softeners, especially cellulose acetate and/or cellulose acetate propionate.

The object mentioned above is further met, according to the invention, in a process of the type mentioned above in that a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm is used as fiber material and the molded body is produced by forming a biodegradable, liquid-tight boundary layer by means of a film coating or foil coating which is formed by applying a foil based on polyester, polyester amide or polylactic acid to the baked molded body.

The boundary layer can preferably be formed as a biological, decomposable, hydrophobic coating which is applied subsequent to the baking of the molded body, e.g., by casting, dipping, or spraying on a solution containing a hydrophobic material such as cellulose acetate or cellulose acetate propionate, wherein it has surprisingly been shown that the use of softeners and, as the case may be, also the use of a binding agent or adhesion promoter can be omitted without prejudicing the adhesion of the coating. The molded body can preferably be coated while already cooled or the coating may also be carried out while the molded body is still hot from the preceding baking process.

The impregnation, e.g., by means of spraying on, ensures even, thin layers and multiple coating can be carried out if need be. According to a further facet of the invention, polyester foil, polyester amide foil or polylactic acid foil is used to form the boundary layer, wherein these coatings are preferably elastic and can preferably be applied (with or without softeners) directly to the molded body without an adhesion promoter by vacuum heat sealing or compression heat sealing.

It was surprising that impregnation with cellulose acetate or cellulose acetate propionate (or a mixture of both of the latter with solvent) had a good adhesion to the molded body without using softeners and, as the case may be, without adhesives while retaining a high fiber content in the molded body. For this purpose, it is essential that the impregnation or coating is sufficiently elastic. Molded bodies of starch and cellulose contain approximately 10% natural moisture which changes along with the humidity of the surrounding air. Changes in the humidity of the surrounding air lead to an expansion or contraction of the molded body. Therefore, in order to ensure that the boundary layer and molded body hold together reliably, the boundary layer must be sufficiently elastic and must conform to the changes in shape of the base material (molded body).

A wide variety of applications is opened up by elastic biodegradable plastic material of the type mentioned above which is free of softeners because softeners constantly migrate out of the polymer composite into the goods, in particular, foodstuff, held in the molded body. The extent of migration depends on the concentration of softener in the contact layer (boundary layer), on the solubility of the softener in the foodstuff and on the use temperature of the associated packaging.

According to the present invention, this problem is completely eliminated because biodegradable plastics material such as cellulose acetate and cellulose acetate propionate which is free of softeners is used as impregnation.

By dispensing with softeners for the production of the molded packaging provided with a hydrophobic boundary layer, this packaging has proven readily usable for boiling water as well as for hot grease.

Depending on the requirements for purposes of use and on the hydrophobic properties, the hydrophobic boundary layer is applied by spraying, casting or dipping also in the form of foils with a thickness of between 20  $\mu\text{m}$  and 200  $\mu\text{m}$ .

The foil material, especially polyester foil, polyester amide foil or, in particular, elastic polylactic acid foil, is selected depending on the purpose of application and the barrier requirements with respect to the melting point and thickness of the foil. The elasticity of the foil in the cold state is essential for a

stable adhesion of the foil to the molded body. While adhesion can be improved by binders, it is preferred that the impregnation or coating is applied to the molded body without the use of a special binder layer. The foil coating is carried out with foils which are free of softeners or which contain biodegradable softeners and are based on the materials mentioned above. The foil can have one or more layers.

According to a preferred embodiment example of the process according to the invention, the foil is heated at its underside which is provided for the adhesive connection with the molded body until close to the melting point of the foil, while the upper side of the foil remains at an appreciably lower temperature.

The foil can be prestretched prior to application to the molded body by means of a male die.

If it is not possible to preheat the foil at its underside, the molded body can also be preheated prior to coating with the foil to a temperature above the melting temperature of the foil and the foil can be applied subsequently by pressure or vacuum.

According to another preferred embodiment form of the process according to the invention, a hydrophobic substance or mixture of hydrophobic substances such as cellulose acetate, cellulose acetate propionate, synthetically or biologically produced and biodegradable polyesters or polyester derivatives, especially polyester amide or polylactic acid, is added to the baking mass in order to render it hydrophobic, so that the baked molded body has its own hydrophobic characteristics at the conclusion of the baking process and, in particular, has a suitable boundary layer which prevents the penetration of liquid in every case for a sufficient period of time.

According to a preferred process for a coating of the baked molded body which forms a barrier against cold or hot liquids at least temporarily, the molded body is initially conditioned with steam, an adhesion promoter is then applied, followed by the coating.

With respect to the preparation of the baking mass, the molded body substantially comprises biodegradable fiber material that is reinforced with starch to

form a fiber-material-and-starch composite action based on the gelatinization of starch.

Depending on the utilized starting materials, i.e., the fiber-containing raw material or the type of biodegradable fibers which can also be used immediately, and on the intended purpose of use as well as on the packaging design (mold depth during the baking process), there is a variable field of process parameters with respect to the mixing ratios of the materials used, namely, water, biodegradable fiber material and starch, and with respect to the utilized comminution equalization and baking technology.

According to a preferred embodiment form of the process according to the invention, this process can be used with a relatively high proportion of fiber material, i.e., waste paper and cellulose fiber-containing and starch-containing production waste or other biodegradable fiber material sources, e.g., beet chips or the like, wherein excellent product properties are nevertheless achieved in the molded bodies produced in this way as packaging materials. The molded bodies or packaging material produced in this way are rapidly biodegradable and can be recycled without great effort. The use of waste paper and cellulose-fiber-containing and starch-containing production residues, e.g., wood pulp and paper pulp, beet chips or the like, leads to a surprisingly high-quality molded body, especially for packaging purposes, which is also extremely durable due to the formation of a fiber material/starch composite and thus elastic and can be produced in a simultaneously economical and environmentally sound manner, even with a relatively small proportion of starch, with a final processing of the equalized viscous mass in a concluding baking process stage using elements of waffle baking technique.

On the other hand, the process according to the invention can also be carried out the appreciably reduced fiber material proportions, preferably up to a ratio of fiber material to starches of 1:4 and up to a fiber material proportion of 10 percent by weight to 30 percent by weight of the viscous mass and with a proportion of 20 percent by weight to 50 percent by weight of modified starch or pregelatinized starch or starch component in the viscous mass (proportion of modified starch or pregelatinized starch in the viscous mass of 1 percent by weight to 13 percent by

weight), wherein molded bodies which are extremely stable with respect to shape are obtained and the fiber proportion is used, especially as raw fiber material, for this purpose.

Preferred configurations of the process according to the invention and compositions of the molded body are indicated in the subclaims.

With respect to the molded body, the object mentioned above is met, according to the invention, in a molded body of the type indicated above in that the molded body has a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length of from 0.5 mm to 50 mm and has, at least on one side, a biodegradable, hydrophobic boundary layer which is formed without softeners on the basis of cellulose acetate or cellulose acetate propionate or which has a fiber coating based on polyester, polyester amide or polylactic acid. This foil coating can be provided with or without softeners.

Ethanol, acetone or ethylene acetate are used as solvents for the impregnation.

The molded body preferably has a porous internal structure which is closed through a peripheral skin layer of greater density, wherein the skin layer carries the liquid-tight coating, preferably without the intermediary of an adhesion promoter layer.

Preferred configurations of the molded body according to the invention are indicated in the rest of the subclaims.

The invention will be described more fully in the following with reference to embodiment examples and the accompanying drawings.

Fig. 1 shows a flow chart of the process for the production of molded bodies based on the use of waste paper as fiber-containing raw material;

Fig. 2 shows a flow chart of another embodiment example for the production of molded bodies based on the use of pre-commminuted, deinked waste paper (deinking material);

Fig. 3 shows a flow chart for another embodiment example of a process for the production of molded bodies based on the use of production wastes as fiber-containing raw material such as paper pulp, beet chips, wood pulp, etc.;

Figs. 4a and 4b show tables for different test samples with different use of native starch and with a constant water fraction in relation to the dry mass of the fiber material;

Figs. 5a and 5b show tables for different test samples with different use of native starch and pregelatinized starch with a constant water fraction in relation to the dry mass of the fiber material;

Fig. 6 shows a table and a corresponding chart showing the grading of the length of the utilized fiber material;

Fig. 7 shows a pressure-time graph for a baking process for producing a molded body according to one of the processes mentioned above;

Fig. 8 shows a product evaluation table for molded bodies as a function of the mold depth of the utilized baking mold and as a function of graded fiber material;

Fig. 9 shows a product evaluation table for molded bodies as a function of the mold depth of the utilized baking mold and as a function of fiber material mixtures with different lengths of fibers and fiber bundles;

Fig. 10 shows a recipe table for the production of molded bodies based on waste paper as fiber-containing material;

Fig. 11 shows a recipe table for making molded bodies, especially for the use of raw fiber material;

Fig. 12 shows an enlarged cross-sectional view of a wall portion of a molded body;

Fig. 13 shows embodiment examples for a hydrophobic impregnation (spray coating) of trays and pots using recipe Y14 (see Figs. 4b, 5b, 11) with cellulose acetate;

Fig. 14 shows embodiment examples for a coating of trays and pots of a molded body according to recipe Y14 (see Figs. 4b, 5b, 11) with cellulose acetate propionate;

Fig. 15 shows embodiment examples for a foil coating of trays and pots with foils based on polyester, polyester amide and polylactic acid;

Fig. 16 shows a product evaluation table for molded bodies coated with cellulose acetate or cellulose acetate propionate with and without softeners;

Figs. 17 to 20 show various embodiment forms of molded bodies which were produced according to embodiment examples of the process according to the invention.

Within the framework of the present Application, the terms fiber material or fiber structure are used in connection with pulping or breaking up to individual fibers and breaking up to only relatively large fiber bundles of the fiber-containing raw material.

Within the framework of the present application, hydrophobic impregnation or liquid-tight coating comprehends the application of the barrier layer whose effect, depending on the material and layer thickness, can also be given only for a determined period of time which may be relatively short. A liquid-resistant characteristic of the molded body which has this characteristic at room temperature (25-40°C) and at a relative humidity of the surrounding air of 0-90% for several days (application class A1) can thus be designated in this way. Further, as a result,

these terms may also be used to designate a resistance to cold water for 24 hours, wherein the packaged goods are at room temperature (25-40°C) or below room temperature (application class A2). Finally, the formation of a biodegradable hydrophobic boundary layer (preferably a coating) also refers to a resistance to boiling water for a period of at least 30 minutes, preferably 1 hour, wherein the packaged goods can have a temperature of 95°C (application class A3). In this connection, the molded body should not lose its stability with respect to shape, and a noticeable penetration of liquid into the molded body structure is prevented.

Within the framework of this patent application, the term hydrophobic impregnation includes the entire hydrophobic range from certain water-repellant or grease-repellant characteristics (no closed border layer, but rather porous coating islands or droplets) to actual water-tightness in the sense of a barrier layer, wherein the latter can sometimes only be achieved by multiple-layer impregnation.

Fig. 1 shows a first embodiment example of the process for producing molded body packaging based on waste paper. However, within the framework of this process, raw material (raw fiber material) can also be used in particular when working with a further reduced fiber fraction in the mass and increased starch fraction (see embodiment example with reference to Figs. 4b, 5b, 11).

Fig. 1 shows, in dashed lines, added materials which may be provided in addition (e.g., pregelatinized starch, fillers, fluxing agents, dye) or the addition of the added materials in alternative stages of the process or in combination with the possibilities shown in solid lines.

In a first process step, the waste paper is comminuted in the dry state, wherein this comminution is preferably carried out in a protective manner without cutting, e.g., by means of shredders, choppers or impact beating mills such as pinned disk mills or hammer mills, so that there is essentially no cutting into the paper fibers, as this leads to a substantially increased water absorption in the continued process due to the resulting increased absorbency of the cellulose fibers of the waste paper. The comminution process should result, through tearing up, in waste paper scraps in an order of magnitude of up to approximately 10 mm, preferably up to approximately 5 mm. For thick-walled and/or large-area molded

bodies, fibers or preferably loosened fiber bundles with a fiber length or fiber bundle length of up to approximately 50 mm can be used to improve inherent stability. Also, for this area of application, a mixture of longer fibers or longer fiber bundles in the range of approximately 10 mm to 50 mm with short fibers or fiber bundles in the range of 0,5 mm to 20 mm can be used.

When longer fiber bundles are used, the preparatory comminution is correspondingly simplified.

As in the other embodiment examples described herein, the preferred separation of the fiber bundles leads to a more intimate connection with the gelatinized starch which can penetrate into the hollow spaces of the fiber bundles so as to promote and enable the formation of a fiber/fiber bundle skeleton (interlaced fibers/fiber bundles) penetrated and superposed by a starch matrix, which leads to an excellent inner binding under the given conditions of a baking process releasing steam under pressure.

The comminuting process is preferably managed in such a way that there is a separation of the fiber bundles of the waste paper scraps affected by the comminution process.

In order to digest or break up its fiber structure, the waste paper which is protectively comminuted and essentially ripped up in the manner described above is then defibrated or pulped by adding water and by softening and further separation or loosening in a mixing and kneading process and is equalized to form a viscous mass.

The mixing and kneading process used for the preferably discontinuous or continuous kneader or combined mixer and kneader is carried out in the presence of water.

Since it is advantageous to maintain the lowest possible water fraction, depending on the rest of the components of the viscous mass and especially depending on the starch proportion (the free water being essentially required for sufficient gelatinization of this starch proportion) in order to be able to carry out the subsequent shaping process (baking) in an efficient manner, it has been shown that a water proportion of 2:1 to 3:1, preferably 2.5:1, in relation to the dry mass of the

waste paper is advantageous for many preferred applications (high-strength molded bodies, dense structure). At the same time, native starch, e.g., native cereal starch, potato starch, corn starch or rice starch, is added to the mass. For protection of the fibers and accordingly for reduced water absorption by the cellulose fibers, the mixing and kneading process for equalizing this viscous mass is effected practically entirely by shearing, i.e., by using shear forces between the mass particles or between the mass particles and a kneading or mixing element, and accordingly by internal friction, so that the waste paper scraps are carefully broken up to their fiber structure.

Within the framework of this patent application, fiber structure comprehends breaking up to individual fibers as well as breaking up to only relatively large, preferably separated or loosened fiber bundles of the starting material.

In many cases it is preferable to carry out the digestion or breaking up only to the fiber bundle since in this way, in combination with the gelatinization of the starch, a fixedly interlaced structure and the formation of a fiber bundle-starch composite are brought about.

The ratio of starch to fiber material in the viscous mass can vary within wide limits and depends in particular on the field of application and the quality requirements for the finished molded body.

For especially smooth surfaces with high elasticity and as a result of the use of loosened fiber bundles which contribute to structural strength to an increased degree, the starch fraction can also be substantially above 50% (in relation to the dry weight of the waste paper), e.g., in a ratio of fiber material to starch of 1:4.

The starch is preferably mixed in as native starch. Fig. 4 shows mixing ratios of native starch to fiber material (waste paper) in relation to the dry mass of the waste paper and while maintaining a 2.5-fold water surplus in relation to the dry mass of waste paper.

The second process step (kneading and mixing process) shown in Fig. 1, in which the waste paper scraps are broken up to their fiber structure (preferably fiber bundles), can be divided into a pretreatment of the waste paper scraps for softening the same by addition of water and further loosening, wherein fluxing agents,

especially ingredients with alkaline effect, for improving the flow properties of the viscous mass in a subsequent baking process as well as filler materials or dyes can also be optionally added at this stage of the process.

Further, it is also possible to premix filler materials with the native starch or portions thereof and to add them in part already during the preceding dry comminution process step or to add the mixture in its entirety during the formation of the viscous mass taking place in the presence of water while carrying out the mixing and kneading process for breaking up the waste paper into its fiber structure.

As will be explained more precisely hereinafter, it is also possible in a particularly advantageous management of the process to use modified starch or pregelatinized starch in addition to the native starch in order to provide an amount of water defined by the pregelatinization state during a subsequent baking process and also to achieve a gelatinization of the starch during very short dwell times or holding times of the mass in the subsequent baking process to form a stable fiber-starch composite. When a pregelatinized starch fraction, whose use is optional, is added as in Fig. 1, the free addition of water can be limited.

Particularly good results with respect to surface quality, elasticity, structural composition, texture and strength were achieved in the produced molded bodies when a mixture of fiber material with fibers of different lengths was also used in addition to a mixture of native starch and pregelatinized starch, especially by using fiber bundles of different length or fiber bundles in combination with individual fibers (when using fiber-containing raw material such as waste paper of different degrees of comminution). Further details will be given in this connection with reference to other embodiment examples (see Fig. 6).

Chalk, kaolin, talcum, gypsum, alumina, titanium oxide or aluminum oxide are taken into consideration in particular as filler materials. Alkaline additions such as magnesium carbonate, magnesium hydroxide carbonate, sodium hydroxide or ammonium hydroxide are used as fluxing agents. The quantity of water added, preferably in a proportion of 2.5-times the dry mass of waste paper material, is that required to achieve a viscous flowing-pulpy to highly-viscous, plastic consistency of

the mass on the one hand and to gelatinize the starch in a subsequent baking process on the other hand.

However, it is also possible to work with a larger amount of surplus water, especially when no pregelatinized starch is used in addition to the native starch. During the thorough mixing and kneading of the viscous mass, the waste paper is broken up or resolved to its fiber structure, preferably to relatively large fiber bundles in this case, and these fiber bundles (or individual fibers) are mixed intimately and in an equalizing manner with the starch, and an intimate connection is made between the softened paper fiber structure and the starch.

Subsequently, after equalizing the viscous mass, this viscous mass is metered in consideration of the subsequent introduction into at least one baking mold and is inserted into the baking mold. The baking mold is formed by at least two baking plates, i.e., an upper baking plate and a lower baking plate (held in baking tongs), wherein the inner surfaces of the baking plates are held at a distance from one another in a closed locked state of the baking mold so as to form a mold cavity, and the mold cavity is filled with the viscous mass. Of course, a plurality of baking tongs can also be used for simultaneous production of a plurality of molded bodies.

Fig. 4a shows the quantities of native starch used for 15 test samples with the amount of water maintained constant (2.5-times) in relation to the dry mass of the waste paper fiber material (fiber bundle).

Fig. 4b shows the quantities of native starch used for 15 other test samples with the amount of water maintained constant (5 times) in relation to the dry mass of the fiber material (fiber bundle), wherein raw fiber material in particular can also be used as fiber material in this case.

With respect to the preferred use of a mixture of native starch and pregelatinized starch, the table in Fig. 5a gives preferred ratios of the use of native starch and pregelatinized starch. It has proven advantageous when the ratio between native starch and pregelatinized starch is approximately 3:1.

In further embodiment examples of test samples, Fig. 5b shows preferred use ratios of native starch and pregelatinized starch. In a total mixture ratio of starch to

fiber material of 1:2 to 4:1, the ratio of native starch to pregelatinized starch can vary between 1:3 and approximately 3:1.

In connection with an increase of the proportion of starch and a reduced proportion of fiber material in the mass (fiber material fraction preferably 10 percent by weight to 30 percent by weight), e.g., with a ratio of fiber material to total starch in the viscous mass (with respect to weight) of 1:4, the ratio of modified starch or pregelatinized starch to total starch is expanded to a range of 1:5 to 1:2. The portion of modified starch or pregelatinized starch in the viscous mass can be increased up to approximately 13 percent by weight.

Further, advantageous characteristics in the molded bodies produced according to the invention were observed when the proportion of starch to water in the viscous mass was approximately 1:10 to 1:1, preferably 1:3 to 1:2.

The baking process whose technique derives from basic elements of waffle baking technology begins with the closing and locking of the baking mold, leaving a mold cavity filled with mass between the upper baking plate and lower baking plate. This distance or gap which is maintained constant by locking the upper and lower baking plates during the baking process determines the wall thickness of the finished molded body.

The baking process, whose progression over time is shown in a pressure-time graph in Fig. 7, is carried out at a temperature between 105°C and 300°C, wherein particularly good results were repeatedly achieved with respect to the thorough baking of the molded body and the geometry of the molded body at a baking temperature of approximately 180°C. The baking period is dependent on various parameters, especially on the viscosity of the mass (water content), the starch proportion (the time period must be such that a complete and thorough gelatinization of the native starch is made possible, also in cooperation with the modified starch or pregelatinized starch that may be used), the geometry of the molded body, and the specific constituents of the mass. In general, the baking period may vary between 0.5 and 15 minutes, wherein shorter cycle periods in the range of 1 to 3 minutes are generally sufficient and result in dimensionally stable molded bodies with a high-quality smooth surface, high elasticity and structural

strength due to the resulting fiber-starch composite or fiber bundle-starch composite of a fiber-reinforced starch matrix.

In other cases, especially with higher proportions of water, a baking period between 3 and 13 minutes has also led to good results, wherein a baking period that is as short as possible is advantageous for increasing the efficiency with which the process is carried out without impairing the uniform inner structural formation of the fiber bundle-starch matrix.

The mold closing force of the baking mold is plotted over time in Fig. 7, wherein the pressure rise during the baking process shown therein to the pressure increase due to the evaporation of and vaporization process of the water in the mass must be considered. It is essential for complete formation of the molded body that a minimum force of approximately 150 kp representing the internal pressure in the mold from the water evaporation measured at the closing pins of the baking mold is not exceeded and that the maximum internal pressure represented by the maximum force occurring at the closing pin, in this case approximately 256 kp, is not substantially exceeded or does not rise excessively.

In Fig. 7,  $t_1$  represents the period from the closing of the baking mold to the time when the value of the mold closing force  $f_{min}$  representing a minimum evaporation pressure in the baking mold is reached,  $t_2$  represents the period from the closing of the baking mold to the time when a value of the mold closing force  $f_{max}$  representing a maximum evaporation pressure inside the baking mold is reached,  $t_3$  represents the period from the closing of the baking mold to the time when steam ceases to be released,  $t_4$  designates the time required for thorough baking or drying of the baked molded body, and  $t_5$  designates the total baking period from the closing of the baking mold to the opening of the baking mold. In the present case, good quality and removability of the molded body from the mold is achieved with a closing force difference  $f_{max}-f_{min}$  of approximately 100 kp, wherein  $t_5$  represents the preferred baking period in the range of 1 to 3 minutes.

The difference  $t_3-t_1$  accordingly refers to the baking process and the shaping accompanied by the release of steam.

During the baking process, the native starch gelatinizes (possibly also accompanied by water absorption by the pregelatinized starch portion) and solidifies. The starch is embedded in the loosened fiber bundle structures of the waste paper and generates a stable binding of the loosened fiber structure with the starch matrix forming by gelatinization.

When only native starch is used, molded bodies of this type are already adequate for a number of applications with respect to elasticity and surface quality. Whereas a high starch proportion leads to a better surface, although with reduced elasticity, the elasticity of the molded body can be reduced through a reduction of the starch fraction, usually only to the detriment of a smooth surface. It is possible, however, by means of the process according to the invention, to achieve a substantial improvement in elasticity while simultaneously ensuring a high surface quality by using modified starch or pregelatinized starch in addition to native starch. The use of a mixture of pregelatinized starch and native starch is especially efficient because there is not sufficient free water available or not a sufficient baking period in the baking process for the gelatinization of the native starch due to the high absorbency of the waste paper. The introduced native starch therefore often does not gelatinize completely in conventional products and results in a wide scatter with respect to the elasticity of the molded body obtained in this way.

According to the invention, characteristics which go substantially beyond the characteristics of conventional molded bodies based on starch can be achieved in that a portion of the starch is pregelatinized with water and the pregelatinized starch is added to the viscous mass, wherein the portions, as was already explained above, are given for a test series from Figs. 4 and 5 (proportion of total starch). The proportions of native starch, pregelatinized starch and water refer in each instance to the dry mass of the waste paper.

Within the framework of the process according to the invention, it has also proven particularly advantageous with respect to surface quality, elasticity, fabric structure and strength of the finished molded body when fiber material of different length, especially fiber bundles of different length or mixtures of longer fiber bundles and shorter individual fibers are used in addition to the native starch and modified or

pregelatinized starch. The fiber length for molded bodies having thin walls is preferably selected in the range of 0.5 to 5 mm, wherein an overview of the fiber fractions (grade) is given in Fig. 6.

In a modification of the process according to Fig. 1, it is also possible, instead of carrying out the comminution of the waste paper in the dry state, possibly with the addition of native starch and filler materials, to carry out the mechanical comminution already in the presence of water in a suitable comminuting mixer.

With respect to the use of a proportion of pregelatinized starch, a modified starch adapted to the baking technology can also be used.

However, for a wide range of uses of the molded body produced by such baking techniques to replace non-biodegradable plastic packaging, e.g., for packaging moisture-containing foodstuff or as disposable take-out packaging for soups, salads and other solid hygroscopic foodstuffs, it is also necessary for the molded body to be sufficiently moisture-resistant without losing its basic biodegradability.

In this connection, it is advantageously provided that the baked molded body is impregnated or coated in a hydrophobizing manner in an inline process, possibly in that the molded body is removed from the baking mold and inserted into another section of the baking device, wherein this can also be carried out outside of the baking device and, depending on the specific coating process, by utilizing the process heat of the baking process for the molded body which is still hot or has already cooled.

Shared driving means and coordinated controlling means are preferably provided between the baking process and subsequent coating process immediately adjacent to the baking mold and within the baking arrangement, possibly with the use of a robot or handling device for removing the baked molded body from the baking mold and for inserting it into a coating device, at least in the case of an inline coating process.

Preferred processes for rendering the baked molded body hydrophobic by applying a hydrophobic impregnation or coating consist in that a spray solution containing cellulose acetate (CA) or cellulose acetate propionate (CAP) or a mixture

thereof as hydrophobic substance is applied (possibly multiple times) to the inside and/or outside of the molded body by spraying, casting or dipping, wherein, depending on the required degree of the hydrophobic characteristics, a water-repellant or fat-repellant application is carried out without the formation of a cohesive boundary layer (e.g., for hamburger packaging) or, when the layer thicknesses are larger, an actual liquid tightness and formation of a continuous boundary layer (barrier layer) can be ensured. Embodiment examples in this connection are shown in Figs. 13 and 14.

In this connection, cellulose acetate is preferably used in the spray solution in a proportion to dry substance of 4.5 percent by weight to 15 percent by weight with acetone as solvent and with a viscosity of the spray solution of between 20 and 4000 mPas, while cellulose acetate propionate, preferably in a weight ratio of 9 percent by weight to 20 percent by weight to dry substance, is used in the spray solution which preferably has a viscosity of 200 to 6,000 mPas, wherein acetone is preferably likewise used as a solvent. Spraying on results in a very even application and accordingly improves hydrophobic protection with thinner layers.

The barrier layers of cellulose acetate and cellulose acetate propionate are applied without softeners and preferably also without adhesion promoters, and it has turned out, surprisingly, that this hydrophobic impregnation has excellent adhesion characteristics even with a relatively high fiber fraction of the molded body. All of the problems which could occur as a result of a migration of softener out of the polymer composite into the packaged foodstuff are prevented from the outset with a coating of this kind.

Of course, it is also possible to apply the impregnation with the intermediary of an adhesive, if needed.

The molded body can be conditioned with steam (increasing the flexibility through application of moisture) concurrently with the hydrophobizing or subsequent to hydrophobization.

The shown in Fig. 11 with reference to other test samples, the process can also be carried out, especially with the use of raw fiber material, with an increased proportion of starch and reduced fiber material fraction with respect to the above-

mentioned CA impregnation and CAP impregnation as well as with respect to the coating processes which will be described hereinafter.

As an alternative to the application of the materials mentioned above (without softeners) as solution coating, outstanding results have also been achieved by coating the molded bodies by films or foils based on polyester, polyester amide or polylactic acid, wherein elastic foil in particular has proven extremely adhesive. A foil coating of this kind can be carried out under pressure or under vacuum, wherein, in the case of deep molds, the foil is preferably prestretched by means of a pressing die and either the foil is preheated on the adhesive side facing the molded body or the molded body which is still warm (utilization of process heat and residual warmth of molded body exiting the baking process) is heated in order to improve the foil adhesion.

These foils can comprise one or more layers and can be used with or without softeners and can also be applied with or without adhesion promoters. In this connection, reference is had to the embodiment examples according to the table in Fig. 15. In practice, the foil coating always results in a liquid-tight coating which also leads in part, especially when using a foil based on polylactic acid (elastic), to resistant characteristics relative to boiling water.

For the above-mentioned spray coating with cellulose acetate and cellulose acetate propionate, Fig. 16 shows conventional solutions (use of a softener with 10 to 30 percent by weight) and the use of CA and CAP, according to the invention, with softeners, wherein the use of cellulose acetate gives a medium adhesiveness, while the use of cellulose acetate propionate surprisingly exhibits outstanding adhesion without hydrophobic softeners.

In an alternative process sequence (process II), a coating of the baked molded body can also be carried out simultaneously in connection with the steam conditioning by spraying on a biodegradable, liquid-tight material. A sterilization of the molded body with hot steam is also preferably carried out subsequently in this case.

In a further embodiment form, the formation of a liquid-tight boundary layer which does not impair the biodegradability of the overall product and a thorough

total impregnation of the molded body can also be carried out in that the utilized material (CA, CAP, polyester, polyester amide, polylactic acid) is introduced into the viscous raw mass as a finely distributed liquid, powder or granulate and is substantially evenly distributed in the molded body material during the baking process, possibly by melting and embedding in the resulting fiber-starch composite and at the conclusion of the baking process leads to a liquid-tight, biodegradable molded body.

The process according to Fig. 1 can therefore also be modified with respect to the application of a liquid-tight coating which follows the baking process in this case such that the material forming a barrier layer against gas-permeability or penetration by liquid is added in a preceding process step already during the preparation of the baking mass and leads to an integrally liquid-tight molded body by rendering the baking mass hydrophobic.

Fig. 12 shows a schematic cross section through a wall portion of the molded body produced according to the process with reference to embodiment example 1. Because of the steam formation and occurring overpressure within the molded body mass during the baking process, a loosened base material layer 20 is formed which is bordered on both sides by a skin layer 21 and in which a starch-fiber material skeleton is formed in connection with a mixture of long and short biodegradable fibers or fiber bundles. The liquid-tight coating 23 adjoins the upper skin layer 21. An adhesion promoter layer can be placed therebetween if desirable. In this case, the molded body is provided with the hydrophobic coating 23 on only one side (e.g., at an inner surface subjected to greater stress). Of course, the hydrophobic coating 23 which encloses the liquid seal of this coating and may be of a different quality with respect to its barrier characteristics can also be formed on both skin layers 21, that is, preferably on the inside and outside of the molded body. The barrier layer or liquid-tight coating 23 can also have different characteristics or thickness on either side of the base material layer 20 or can comprise different hydrophobic materials.

The hydrophobic impregnation or coating can preferably also be carried out by dipping the molded body in the coating material or by spraying on a hydrophobic solution with cellulose acetate or cellulose acetate propionate. With respect to the

foil coating according to Fig. 15, application is carried out by adhesion of the foil under vacuum, by means of compressed air or by mechanically pressing on, wherein excess foil overlapping the edges can be eliminated in conjunction with cleaning the baked molded body of material residues proceeding from the exhaust steam channels.

If necessary, in order to form a hydrophobic, especially a liquid-tight boundary layer only on the inside of the molded body, a melt of one of the hydrophobic materials mentioned above can also be applied to a molded article located in the baking mold and the molded body can be baked while forming the boundary layer.

The thickness of the liquid-tight biodegradable boundary layer is preferably in the range of 5  $\mu\text{m}$  to 200  $\mu\text{m}$  when using foils in the range of 20  $\mu\text{m}$  to 200  $\mu\text{m}$ , for example.

Depending on the desired barrier effect and resistance or durability, the hydrophobic impregnation or foil coating can also be applied as a multiple coating, wherein either the liquid-tight boundary layer which is formed so as to be integral by rendering the baking mass hydrophobic or the liquid-tight boundary layer that is formed as a coating applied after the baking process has a barrier effect relative to liquids, especially relative to water, acidic foodstuffs, lactic acid or fats. If necessary, the coating can also be gastight, especially resistant to permeation by steam or oxygen.

With respect to the foil coating with foil based on polyester, polyester amide or polylactic acid, this foil can be taken off from rolls or sheets. The foil can be applied to the molded body which is still hot from the baking process using an adhesion promoter if required or may possibly be applied by melting on or can be laminated onto the cooled molded body under vacuum, by means of compressed air or by pressing on mechanically.

In other embodiment examples, the hydrophobic material can be in the form of powder or granulate or pellets and the molded body can be provided with the hydrophobic coating by powdering on, spraying on or dipping, or a hydrophobization can be carried out directly by introducing these materials into the baking mass

before the baking process as an alternative to the subsequent coating of the molded body shown in Fig. 1 for formation of a liquid-tight boundary layer. The material is preferably melted on and the molded body is still hot from the preceding baking process.

It is further advantageous to increase the surface density of the coated molded body by reducing the porosity for achieving a smooth surface, e.g., by means of a subsequent (precision) pressing process in order to further increase the barrier effect of the coating, e.g., against hot water, hot grease or the like. When using an adhesion promoter between the surface of the molded body and the liquid-tight coating, this adhesion promoter can preferably comprise nitrocellulose or polyvinyl alcohol and has, in the latter case, a gas-blocking action at the same time.

In the case of concave molded body packaging, e.g., in the form of a dish or platter, the hydrophobic coating or formation as boundary layer is provided at least on the inside, but may also be provided externally, wherein the inner surface of the molded body packaging may possibly be subjected to stricter requirements for the barrier effect of its surface layer with respect to resistance to temperature and grease than the outer surface for which, in particular, resistance to moisture and climate may be adequate for a sufficient period of time.

In another embodiment form of the hydrophobic impregnation following the baking process, material is preferably applied to the baked molded body as an emulsion or solution, wherein the layer is cured or hardened after application and may be treated by means of special after-heating treatment.

In addition to the hydrophobic substances cellulose acetate and/or cellulose acetate propionate, the solution preferably contains a volatile solvent. For example, a cold solution containing acetone or ethyl acetate as solvent can be sprayed onto the preferably cooled molded body or this molded body can be dipped in the solution.

Depending on the purpose for which it is used and on corresponding application classes, the inner and/or outer liquid-tight coating of the molded body is carried out or the formation of a hydrophobic characteristic of the molded body is ensured by the liquid-tight boundary layer during the baking process by prior

admixing. In this connection, the application classes range from the lowest requirement stage A1 (room temperature 25-40°C, relative air humidity of 0-90%), barrier effect of the hydrophobic boundary layer over several days, to application class A2, resistance to cold water also at room temperature (25-40°C) for 24 hours, and to application class A3, resistance to boiling water at 95°C guaranteed for up to one hour. For the individual application classes, the coating material also varies with respect to layer thickness and applied concentration.

The lowest application class A1 is taken into consideration particularly for an outer coating of the molded body when this molded body can be closed by a cover at the same time or for coating the inside and the outside when the molded body packaging remains open. Depending on the purpose of use, application class A2 (cold-water-resistant) or A3 (resistant to boiling water) is provided, wherein the outer coating can have a correspondingly lower application class or the same application classes with respect to coating quality.

Moisture absorption of the molded body can also be reduced in that a substantially denser skin layer (compare the schematic view in Fig. 12) is achieved as a result of the baking process by suitable use of separating or parting agents (fully hydrogenated fat or wax) during the baking process.

Before applying a hydrophobic coating to the baked molded body, this molded body is preferably subjected to a priming or filling with a filler material in order to close the pore structure of the baked molded body, wherein, e.g., waxes, polymers, or lubricants can be used as filler materials and this filling serves at the same time as an adhesion promoter for the adhesive bonding of the hydrophobic layer, wherein the adhesion promoter layer can likewise be applied, for example, by dipping, spraying on and/or heating.

For a higher application class (e.g., A3), the hydrophobic coating is carried out with a higher concentration (e.g., 50%) and is also applied repeatedly (e.g., three times). For lower requirements with respect to the barrier effect in the same application class (e.g., A3), lower concentrations are also sufficient and a multiple application can usually be dispensed with.

The formation of the hydrophobic, possibly liquid-tight boundary layer by hydrophobization of the baking mass or application of the hydrophobic coating must at the same time allow for the sealing capability (cover sealing) and should also take into account the furnishing of the molded body with color and printing in an uncomplicated manner.

An indicator for moisture and/or time is preferably provided on the outside of the molded body in order to show the barrier effect provided over the life of the product.

With respect to the biodegradability of the molded body, which is also not impaired through the hydrophobic boundary layer, it can be advantageous to comminute the packaging body after use in order to further accelerate the biodegradability of the molded body.

If required, the molded bodies can be enclosed by a shrink foil comprising one of the aforementioned biodegradable materials as additional protection for shipment.

A further embodiment example of the process according to the invention is described hereinafter with reference to Fig. 2.

In this case, deinking material, i.e., deinked waste paper whose state already ranges from pulverized to small scraps, is used as starting material (fiber-containing raw material containing biodegradable fibers, especially a material of plant origin containing cellulose fibers), although this process can also be carried out (with the inclusion of a comminution step) with waste paper as raw material as in the process according to Fig. 1 or with other fiber material (raw material) containing biodegradable fibers.

In this process, the deinking material (or raw material) which is already sufficiently comminuted is premixed in the dry or moist state with a portion of native starch, possibly, as in the process according to Fig. 1, with the addition of filler materials (which can also be added as a premix with the native starch) and possibly with the addition of fluxing agents of the type mentioned above and/or dyes, while another portion of native starch is pregelatinized with water. The dry premixing facilitates good equalization and homogenization of the mass and is followed by the

further process step consisting of mixing and kneading in the presence of water for breaking up the deinking material to its fiber structure, i.e., preferably to loosened fiber bundles for forming an equalized viscous mass which is added in addition to the portion of pregelatinized starch taking into account the mixing ratios of water, fiber material, native starch and pregelatinized starch indicated in connection with the process according to Fig. 1 and Figs. 4a, 4b; 5a, 5b, Fig. 10 and Fig. 11. As was already demonstrated in connection with the embodiment example, a hydrophobization of the material and formation of the hydrophobic boundary layer can also be carried out already during the baking process especially by addition to the dry premix.

If required, fluxing agents can also be added in during the mixing and kneading process which takes place in the presence of water; this also applies to the addition of filler material or dye suspensions. In this case also, the deinking material is broken down to its fiber structure in the most protective manner possible without cutting, especially by forming relatively large, loosened fiber bundles, wherein improved molded body characteristics can be achieved by directed use of graded materials, i.e., fiber bundle mixtures of different fiber length.

The dry mixing of the waste paper and/or deinking material components (or other cellulose-containing and starch-containing production waste as fiber-containing raw material, see process according to Fig. 3, or also raw fiber material) with starch leads to an improvement in the surface quality of the molded body.

The rest of the process steps, namely, metering and the baking process (possibly including prior shaping), correspond to the concluding process steps described already with reference to Fig. 1 for production of the molded bodies, especially for packaging purposes, using baking techniques and the subsequent hydrophobic coating of the molded body in the manner already described.

As was already described in connection with the process according to Fig. 1, the elasticity of the fully baked molded body is determined to a decisive extent by the use of a mixture of pregelatinized starch and native starch, especially by the pregelatinized portion. Reference is had to Figs. 4a, 4b; and Figs. 5a, 5b, Fig. 10,

Fig. 11 with respect to the quantity ratios which have been shown to be advantageous.

The surface of the molded body is also determined in part by the steam pressure, discussed with reference to Fig. 7, present in the baking mold during the baking process (represented by the mold closing pressure of the baking mold). This steam pressure is dependent on the proportion of native starch and on the moisture of the viscous mass.

The steam pressure in the baking mold can be controlled in the desired manner by methods ranging from control of the steam discharge channels in the baking mold with respect to cross section and position to the use of control valves.

If required, the further addition of water to the mixing and kneading process for producing the viscous mass (shown in dashes in Fig. 2) can be dispensed with entirely, so that the moisture of the mass and accordingly also the proportion of water for pregelatinization of the native starch is determined by the proportion of water in the pregelatinized starch that is used. In this way, the steam pressure ratios during the baking process and thus the surface quality of the molded bodies can also be monitored. The native starch is preferably added in the mixing process with the other constituents of the dry mass, but can also be introduced, at least in part, directly into the mixing and kneading process which breaks down the fiber bundles and equalizes the viscous mass (also shown in dashes in Fig. 2). In the present case, these further constituents are dry comminuted waste paper and/or deinking material or, alternatively, additional fluxing agents and/or filler materials.

The structure and accordingly also the strength of the molded bodies is substantially determined by the ratio of starch to waste paper or deinking material as well as by the length of the utilized fiber bundles or fibers which essentially results in different strengths as has been shown by analysis of comparison models illustrated in Figs. 8 and 9.

It has been shown in particular that the use of fiber-containing material with biodegradable fibers of different length, i.e., fiber bundles or fiber mixtures with different fiber lengths ranging from 0.5 mm to approximately 10 mm, especially for thin-walled molded bodies, preferably ranging from 1 mm to 5 mm, yields very

advantageous results with respect to strength taking into account different molded body geometries and mold depths of the baking mold.

It has been shown that a mixture of short and long fibers or short and long fiber bundles or a mixture of long fiber bundles and short fibers in connection with the use in the baking process of modified starch or pregelatinized starch and native starch can increase the strength of the molded bodies to a surprisingly high degree (see Fig. 9).

In general, it has been determined that a good surface, but only relatively low elasticity, can be achieved in the molded body with a high proportion of native starch in relation to pregelatinized or modified starch, whereas a very high proportion of pregelatinized starch or modified starch in relation to native starch leads to good elasticity, but a poorer surface of the molded body.

Therefore, as regards the respective application and molded body geometry, the ratio of native starch to pregelatinized starch (preferably 3:1) should be adapted in consideration of the fiber lengths of the fiber-containing material or of the mixture of fibers or fiber bundles of different length, wherein especially advantageous ratios have been shown in molded bodies whose composition is shown in Figs. 10 and 11, also with respect to a relatively short baking process of between 0.5 and 3 minutes at approximately 150°C to 200°C.

A further embodiment example of the process according to the invention is shown in Fig. 3, wherein cellulose fibers, paper pulp, beet chips and wood pulp (cellulose fibers) are selected as starting material as representative examples of production waste, wherein other production waste, especially that based on cellulose, containing biodegradable fibers can also be used for this purpose.

Of course, biodegradable fiber structures (raw fiber material) which have already been broken up directly, especially fiber bundles with a length between 0.5 mm and approximately 50 mm (higher for large-area and/or thick-walled molded body packaging), preferably between 1 mm and 5 mm (especially for thin-walled, light molded body packaging), can also be used within the framework of the process according to the invention.

Fig. 3 shows that the process can also be carried out using mixed raw materials from the paper industry, foodstuff industry, feedstuff industry or various materials recycled from waste paper, wherein in Fig. 3 these materials are already sufficiently comminuted, especially by using shredders, choppers or impact beating mills such as pinned disk mills or hammer mills, i.e., as far as possible by tearing and non-cutting processing for protection of the fibers (and for reducing the water absorption of cellulose fibers), wherein the comminuted raw material is graded or classed in a further process step initially with respect to the existing fiber length and its starch proportion for adjusting the respective additional components (addition of native starch and pregelatinized starch or selection of mixing ratios of long and short fibers or fiber bundles).

The rest of the process steps using preferred dry mixing of fiber-containing raw material and native starch and possibly fluxing agents and/or filler materials (insofar as they are in the dry state) and for forming a liquid-tight boundary layer correspond to those that have already been described above with reference to the process according to Figs. 1 and 2. Although a dry premixing between comminution and plasticizing is preferred for the viscous mass, the materials used for the viscous mass can also be combined directly in the mixing and kneading process, possibly in the presence of water.

However, it has been shown that particularly advantageous results are achieved when the mixing and kneading process is carried out without the addition of water merely with the addition of moisture due to the starch which is pregelatinized by excess water, wherein this results in a highly viscous, pasty mass. During the baking process, vegetable water inherent in starch as well as the excess water resulting from the pregelatinized starch is available for gelatinizing the native starch. Molded bodies of especially advantageous quality with respect to surface, texture, elasticity, structure, strength and stability with respect to the mold depth of the baking mold or molded body depth result with the parameters shown in Figs. 10 and 11, wherein the elasticity and the structure are substantially determined in part by the use of pregelatinized starch. The surface and framework structure are further substantially determined by the released steam conditions during the baking

process, i.e., by the moisture fraction of the viscous mass, which should be as low as possible, wherein it is sufficient in many cases for preparing the mass to limit the moistening to the indirect addition of water in the form of admixing with excess water of pregelatinized starch. This moisture is sufficient for bringing about complete gelatinization of the native starch.

A further substantial influence is the length of the utilized loosened fiber bundles, possibly also in connection with individual fibers, wherein the use of fibers also essentially determines the flow behavior of the mass in the baking mold and must be selected while taking into account the geometry and especially the depth of the molded body, wherein the use of short and long fibers in the mixture (long and short fiber bundles) has proven superior in many cases to the use of graded fiber bundles of relatively fixed length in the range of 0.5 mm to 5 mm, as a comparison of Figs. 8 and 9 will show. However, longer fibers/fiber bundles of up to approximately 10 mm can also be used depending on the requirements for the molded body, wherein this also reduces the requirements for the preceding comminution process, e.g., of the waste paper. Of course, although this is not shown in the process according to Figs. 1 and 2, when mixtures of fibers and fiber bundles of different length are used, the fiber material can be graded according to the length of the fibers/fiber bundles or with respect to starch content for evaluation of the starting material.

When using moist deinking materials, for example, the dry mixing process step in the process according to Figs. 2 and 3 can also be carried out in the moist state, possibly also with a slight addition of water.

It can also be advantageous to add viscous mass to the mold after it has already been closed at the beginning of the baking process in order to improve the mold filling behavior in the case of a pasty mass and to work with a slight after-pressing of the upper baking plate against the lower baking plate during the baking process rather than with mold halves which are rigidly closed. However, it has generally proven advisable to adhere to the baking techniques with rigidly closed baking plates (constant distance = wall thickness of molded body during the baking process).

If required, the baking process according to the process shown in Figs. 1 to 3 can be followed by a conditioning of the obtained molded body removed from the baking mold, especially by the application (spraying on) of the liquid-tight material as a solution, emulsion or suspension in a steam atmosphere.

A comparison of Figs. 8 and 9 again shows the advantages of the use of fiber mixtures of different fiber lengths compared with the use of cellulose fibers or fiber bundles which only represent a determined fiber length or a fiber length within a narrow range.

Preferred recipes for the configuration of the process according to the invention are shown in Figs. 10 and 11. It will be seen that it was also possible to produce molded bodies with a very large depth having good strength, elasticity, structure and surface structure by the process according to the invention when the fiber material used, especially loosened fiber bundles, in relation to the total viscous mass was 15 to 30 percent by weight and the total proportion of starch (native starch plus pregelatinized starch) in relation to the total viscous mass was approximately 40 to 5 percent by weight, and the proportion of water in the total viscous mass was approximately 45 to 70 percent by weight and the proportion of pregelatinized starch in the total viscous mass was approximately 10 to 1%.

On the other hand, very good results were also achieved with more starch materials and less fiber materials (especially when using raw fiber) with coatings according to Fig. 11 (increased proportion of pregelatinized starch up to 13 percent by weight).

Embodiment examples of molded bodies which were produced by the process according to the invention are shown in Figs. 17 to 20. Fig. 11 shows molded body packaging 1 with a base part 2 and cover part 3 which are connected by a hinge 4. This molded body packaging 1 is suitable for holding foodstuff, also hot foodstuff, in a closable manner. The liquid-tight boundary layer is not shown more in more detail herein. The molded body packaging 1 has at least one inner hydrophobic boundary layer, preferably a coating of application phase A2.

Fig. 18 shows a molded body (half shell) which can be connected with a corresponding complementary molded body to form a completely closed package.

The shell-shaped molded body 1 has a plurality of depressions 5 in the shape of cylinder sections, wherein these depressions are divided into a longer portion 6 and a shorter portion 7 and this arrangement is repeated in a symmetrical manner on the other side of the molded body so as to be separated by a center web 8. Lateral feet 9 increase stability with respect to shape and improve the support and stacking ability of the molded body packaging. A packaging shell of this kind can be used, e.g., for receiving ball-point pens, lipsticks, cosmetics, pencils, writing instruments or can also be used in pharmacies, e.g., for holding packaging tubes. In this case, a sufficient resistance of the molded body packaging 1 against climatic environmental influences (moisture) - application class A1 - is generally adequate.

Figs. 19 and 20 show relatively deep packaging bodies 1 such as can be used, e.g., as flowerpots, seedling trays or for other packaging or wrapping purposes.

All of the molded body packaging 1 produced according to the process described above could be manufactured quickly, economically and with excellent material characteristics with respect to stability of shape, breaking strength and elasticity, structural density and surface quality and, although biodegradable, are subjected to a hydrophobizing treatment, i.e., impregnation with cellulose acetate and/or cellulose propionate or a foil coating with foil based on polyester, polyester amide or polylactic acid.

Patent Claims:

1. Process for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material, water, and starch and is baked in a baking mold so as to form a fiber material-starch composite, characterized in that a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm is used as fiber material, and the molded body is produced by impregnation with a biodegradable, hydrophobic boundary layer, wherein the boundary layer is formed from cellulose acetate and/or cellulose acetate propionate without softeners.
2. Process for producing molded bodies with a barrier layer, especially molded bodies for packaging, from biodegradable material using a viscous mass which contains biodegradable fiber material and starch and is baked in a baking mold so as to form a fiber material-starch composite, characterized in that a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length in the range of 0.5 mm to 50 mm is used as fiber material and the molded body is produced by means of a foil coating formed by applying a foil based on polyester, polyester amide or polylactic acid to the baked molded body so as to form a biodegradable, liquid-tight boundary layer.
3. Process according to claim 2, characterized in that the biodegradable boundary layer is formed without softeners.
4. Process according to claim 2 or 3, characterized in that the foil has a thickness of between 20  $\mu\text{m}$  and 200  $\mu\text{m}$ .
5. Process according to claim 1, characterized in that the impregnation is carried out by spraying on, casting or dipping with a hydrophobic solution.

6. Process according to at least one of the preceding claims 2 to 4, characterized in that the foil coating is carried out by vacuum heat sealing or compression heat sealing and/or with heat treatment.

7. Process according to at least one of the preceding claims 1 to 6, characterized in that the impregnation or foil coating is elastic and is preferably applied directly to the molded body without adhesion promoters.

8. Process according to at least one of the preceding claims 2 to 4, 6 or 7, characterized in that the foil is heated at its underside intended for adhesive connection with the molded body.

9. Process according to at least one of the preceding claims 2 to 4, 6 to 8, characterized in that the foil is prestretched prior to application to the molded body by means of a male die, especially for hollow molded bodies.

10. Process according to at least one of the preceding claims 2 to 4, 6 to 9, characterized in that at least the adhesion surface of the molded body to be coated with foil is preheated prior to coating with foil to the melting temperature of the foil and the foil is then applied thereto.

11. Process according to at least one of the preceding claims 1 to 10, characterized in that the hydrophobic boundary layer is formed by impregnation or a liquid-tight barrier layer by applying a foil to the molded body when it is still hot from the preceding baking process and the molded body is conditioned simultaneously or subsequently.

12. Process according to at least one of the preceding claims 1 to 11, characterized in that a thickness of the hydrophobic, biodegradable boundary layer is 5 µm to 200 µm.

13. Process according to at least one of the preceding claims 1 to 12, characterized in that the molded body is provided with the boundary layer on the inside and/or on the outside.
14. Process according to claims 1 to 5, 7, or 11 to 13, characterized in that the impregnation is applied as a multiple layer.
15. Process according to at least one of the preceding claims 1 to 14, characterized in that the boundary layer serving as barrier layer has a barrier effect against liquids, especially water, acidic foodstuffs and fats.
16. Process according to claim 15, characterized in that the boundary layer forms a substantially liquid-tight and gastight barrier layer against steam, oxygen and aromas.
17. Process according to at least one of the preceding claims 1 to 16, characterized in that an inner boundary layer of the molded body has a greater resistance to moisture and/or temperature and/or fats and/or is more tightly sealed against gas and aromas than an outer surface of the molded body, especially an outer surface provided with a hydrophobic boundary layer.
18. Process according to at least one of the preceding claims 1 to 17, characterized in that the molded body is provided along an inner surface with a hydrophobic impregnation or coating which is thicker than that on its outer surface.
19. Process according to at least one of the preceding claims 1 to 18, characterized in that the molded body is baked in the baking mold, subsequently removed from the baking mold and introduced into a further device within the baking apparatus for applying the hydrophobic impregnation or foil coating.

20. Process according to at least one of the preceding claims 1 to 19, characterized in that for subsequent impregnation or coating of the molded body, use is made of process heat of the baking mold, preferably within the baking mold, or residual heat of the molded article from the preceding baking process to heat the molded body and/or to heat a hydrophobic medium, especially a coating foil, to be applied to the molded body.

21. Process according to at least one of the preceding claims 1, 5, 7 or 11 to 20, characterized in that a solution containing cellulose acetate and/or cellulose acetate propionate as hydrophobic substance and a solvent, especially a volatile solvent, is used for impregnation.

22. Process according to at least one of the preceding claims 2 to 4, 6 to 10, characterized in that the foil is trimmed at the coated molded body after the molded body is removed from the baking mold in conjunction with a severing of overlapping material resulting from the steam discharge ducts of the baking mold.

23. Process according to at least one of claims 1 to 23, characterized in that fiber-containing raw material which is pulped or defibrated in particular by prior comminution is used to form the fiber material.

24. Process according to at least one of the preceding claims 1 to 23, characterized in that the fiber-containing raw material is waste paper, recycled material, especially deinked waste paper, biodegradable fiber material such as production waste containing cellulose fibers, especially wood pulp or paper pulp, beet chips or the like.

25. Process according to at least one of the preceding claims 1 to 24, characterized in that the fiber material is formed directly from biodegradable fibers or fiber bundles.

26. Process according to claim 25, characterized in that the fibers or fiber bundles have a length ranging from 0.5 mm to 5 mm.
27. Process according to at least one of the preceding claims 1 to 25, characterized in that the starch used is native starch and/or pregelatinized starch or modified starch.
28. Process according to at least one of the preceding claims 1 to 27, characterized in that the ratio of starch to fiber material in the viscous mass is in the range from 15 percent by weight to 400 percent by weight in relation to the dry weight of the fiber-containing raw material, especially waste paper.
29. Process according to at least one of the preceding claims 1 to 28, characterized in that water is added in a ratio of approximately 8:10, preferably 2.5:1, in relation to the dry mass of the fiber-containing raw material, especially waste paper, for forming the viscous mass.
30. Process according to at least one of the preceding claims 27 to 29, characterized in that the proportion of pregelatinized starch or modified starch of the total starch in the viscous mass is approximately 20 to 75 percent by weight.
31. Process according to at least one of the preceding claims 1 to 30, characterized in that the proportion of starch to water in the viscous mass is approximately 1:10 to 1:1, preferably 1:3 to 1:2, and water is subsequently added in the form of modified starch or starch which is pregelatinized accompanied by excess water to form the viscous mass.
32. Process according to at least one of the preceding claims 1 to 31, characterized in that the portion of fiber material in the viscous mass is 10 percent by weight to 30 percent by weight, the proportion of starch in the viscous mass is 5

percent by weight to 40 percent by weight, and the proportion of water in the viscous mass is 70 percent by weight to 40 percent by weight.

33. Process according to at least one of the preceding claims 27 to 32, characterized in that the proportion of pregelatinized starch or modified starch in the viscous mass is 1 percent by weight to 13 percent by weight.

34. Process according to at least one of the preceding claims 1 to 33, characterized in that the fiber-containing raw material, especially waste paper, is comminuted and subsequently broken down to its fiber structure in the presence of water and with the addition of starch, preferably native starch, and the moldable viscous mass is formed and subsequently baked to form molded bodies.

35. Process according to at least one of the preceding claims 1 to 33, characterized in that the fiber-containing raw material, especially waste paper, is comminuted in the presence of water and is subsequently broken down to its fiber structure with the addition of starch, preferably native starch, and the moldable viscous mass is formed and subsequently baked to form molded bodies.

36. Process according to at least one of the preceding claims 1 to 35, characterized in that the viscous mass is metered prior to baking.

37. Process according to at least one of the preceding claims 1 to 36, characterized in that the fiber-containing raw material or the fiber material that is used directly is analyzed with respect to its fiber length and starch proportion and is graded prior to or after the comminuting process.

38. Process according to at least one of the preceding claims 1 to 37, characterized in that the native starch is added in partially already during a comminution of the fiber-containing raw material, especially as a premix with filler materials.

39. Process according to at least one of the preceding claims 1 to 40, characterized in that the starch is added to the fiber-containing raw material, at least partially, during the comminuting process and/or during a subsequent drying process or wet mixing process and/or during a subsequent equalizing mixing and kneading process as native starch and/or modified starch and/or pregelatinized starch.

40. Process according to at least one of the preceding claims 27 to 39, characterized in that the starch is introduced as pregelatinized starch and as native starch and at least a filler material is added into the native starch prior to adding in the comminuting process of the fiber-containing raw material or in the subsequent equalizing mixing and kneading process.

41. Process according to at least one of the preceding claims 27 to 40, characterized in that the pregelatinized starch is added directly to the mixing and kneading process for forming the viscous mass.

42. Process according to at least one of the preceding claims 1 to 41, characterized in that the formation of the viscous mass is carried out using fiber-containing raw material of different grades of comminution and different fiber lengths.

43. Process according to at least one of the preceding claims 1 to 42, characterized in that the mass is heated before introducing it into the baking mold.

44. Process according to at least one of the preceding claims 1 to 43, characterized in that the at least two baking plates of the baking mold are locked relative to one another at the beginning of the baking process while ensuring a constant distance between the upper baking plate and lower baking plate during the baking process, wherein the distance between the inner surfaces of the baking

plates which face one another and between which the mass is located determines the wall thickness of the baked molded body.

45. Process according to at least one of the preceding claims 1 to 44, characterized in that a controlled discharge of steam from the baking mold is carried out during the baking process.

46. Process according to claim 45, characterized in that the discharge of steam from the mold cavity of the baking mold is carried out so as to be controlled with respect to time and/or location and/or internal pressure.

47. Process according to at least one of the preceding claims 1 to 46, characterized in that long fibers or long fiber bundles with a length of up to 50 mm are preferably used for thick-walled and/or large-area molded bodies, especially fibers or fiber bundles with a fiber length or fiber bundle length in the range of 10 mm to 50 mm in a mixture with short fibers or short fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 20 mm.

48. Molded body, especially molded body for packaging, made from biodegradable material, comprising biodegradable fiber material, starch and a residual proportion of water, especially produced in accordance with the process according to at least one of the preceding claims 1 to 47, characterized in that the molded body has a mixture of long and short fibers or fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 50 mm, and has, on at least one side, a biodegradable hydrophobic boundary layer which is formed based on cellulose acetate or cellulose acetate propionate without softeners or has a coating foil based on polyester, polyester amide or polylactic acid.

49. Molded body according to claim 48, characterized in that the molded body is covered on all sides.

50. Molded body according to claim 49, characterized in that long fibers or long fiber bundles with a length of up to 50 mm are preferably used for thick-walled and/or large-area molded bodies, especially fibers or fiber bundles with a fiber length or fiber bundle length in the range of 10 mm to 50 mm in a mixture with short fibers or short fiber bundles with a fiber length or fiber bundle length of 0.5 mm to 20 mm.

51. Molded body according to at least one of the preceding claims 48 to 50, characterized in that the proportion of fiber material to starch is 4:1 to 1:4.

52. Molded body according to at least one of the preceding claims 48 to 51, characterized in that the molded body contains a proportion of pregelatinized starch or modified starch.

53. Molded body according to at least one of the preceding claims 48 to 51, characterized in that the molded body contains a mixture of, in particular, long, loosened fiber bundles and, in particular, short individual fibers or fiber bundles.

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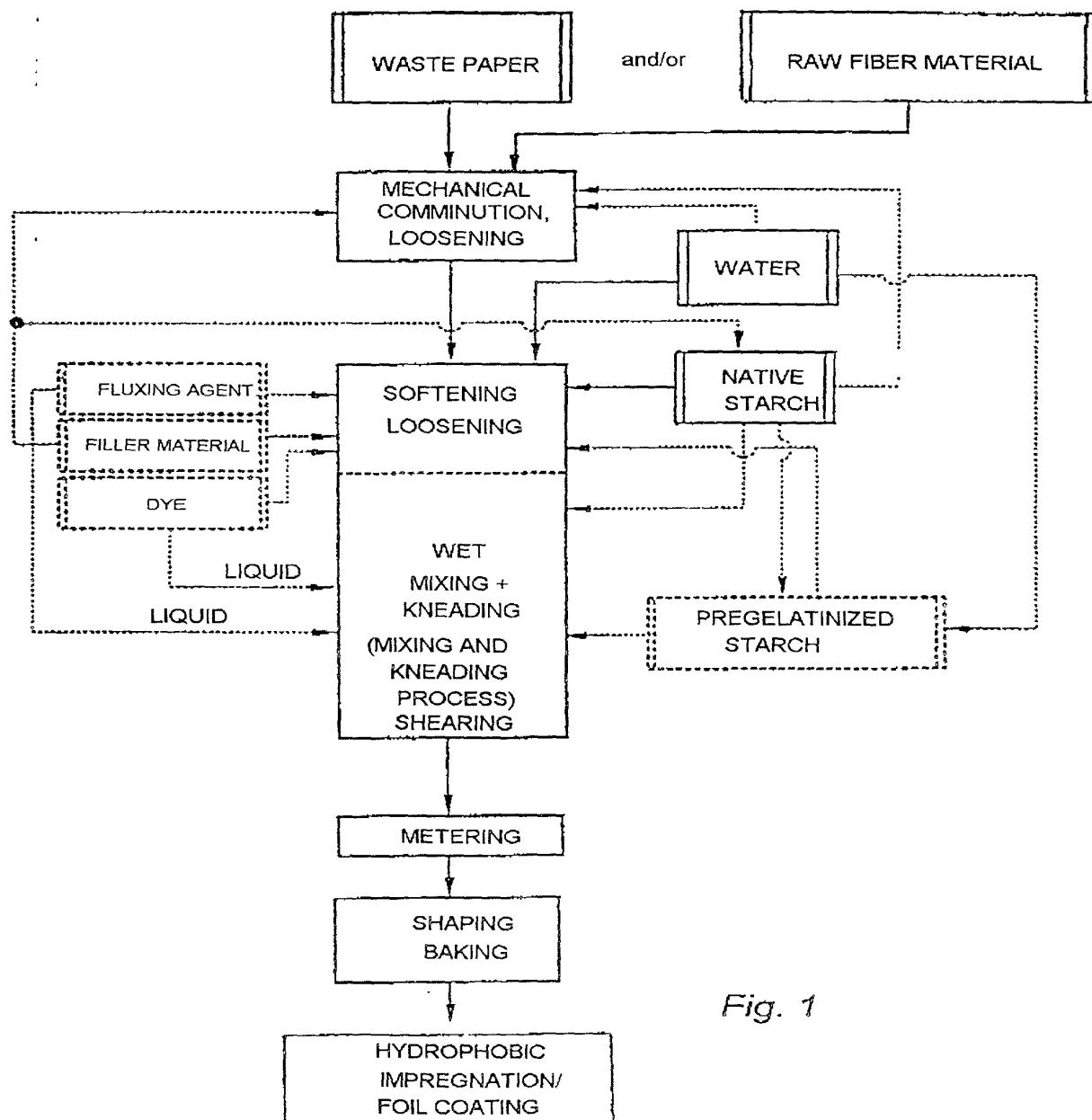


Fig. 1

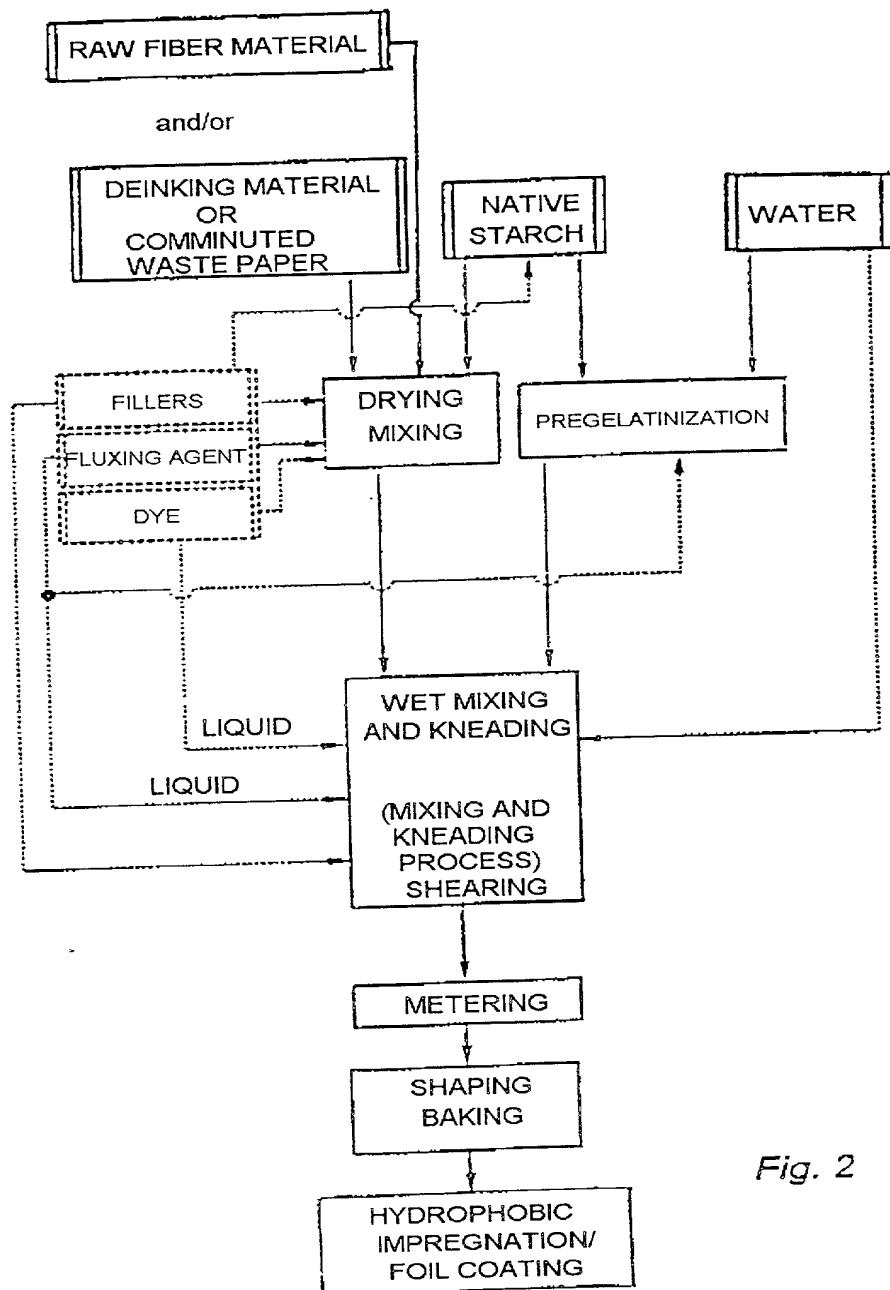


Fig. 2

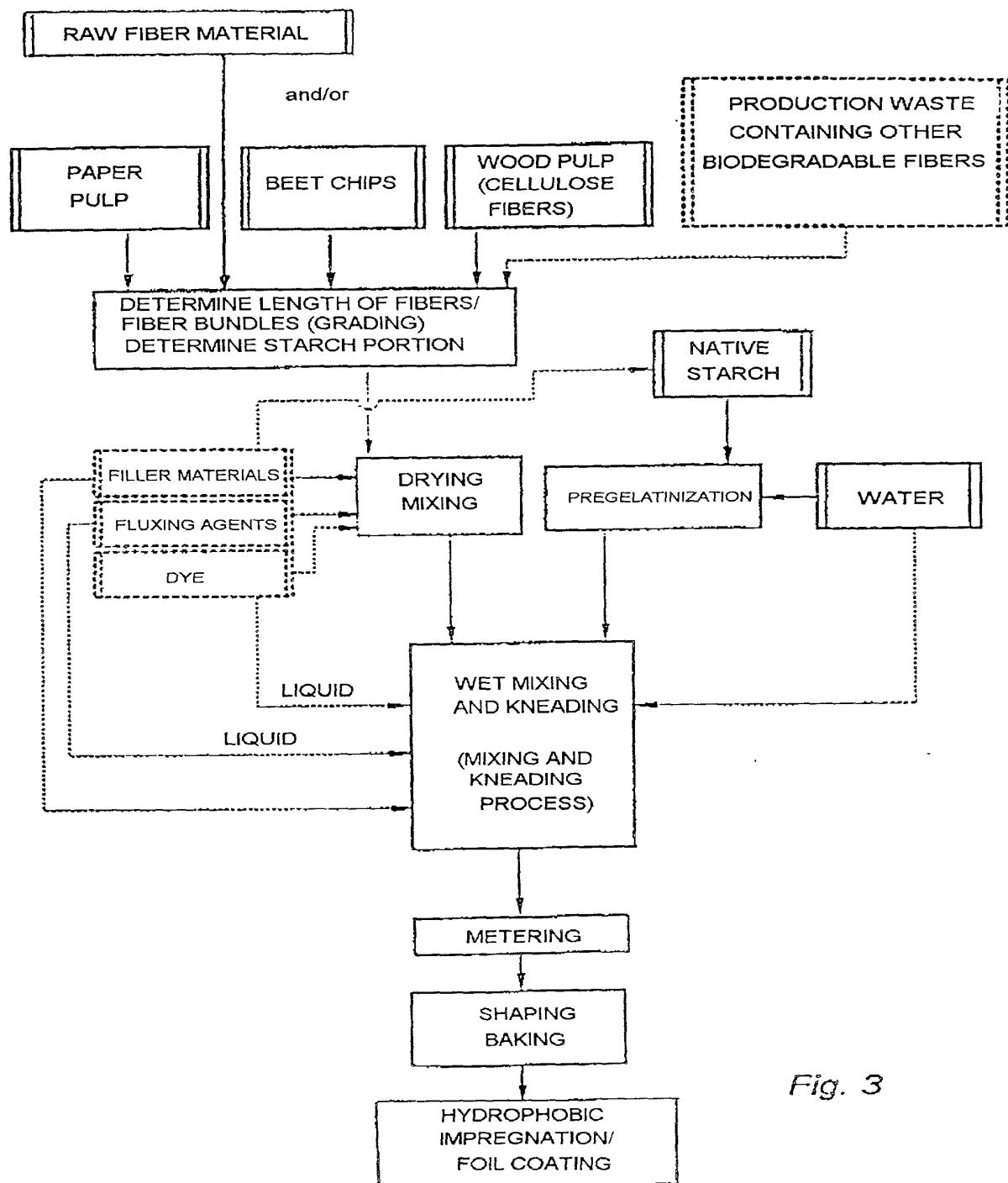


Fig. 3

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	$x_1$	$x_2$	$x_3$	$x_4$	$x_5$	$x_6$	$x_7$	$x_8$	$x_9$	$x_{10}$	$x_{11}$	$x_{12}$	$x_{13}$	$x_{14}$	$x_{15}$
a	18,7	28,2	37,1	47	56,4	65,8	75	84,1	93,3	102,9	112,3	122	131,4	140,8	150

 $x_{1-15}$  test sample

a in wt. % native starch

b = 250 wt. % water (in relation to dry mass  
of fiber material)

c = 100 wt. % fiber material

Fig. 4a

## native starch

Prozentmuster	y1	y2	y3	y4	y5	y6	y7	y8	y9	y10	y11	y12	y13	y14	y15
a	25	64	111	131	150	169	166	206	225	244	281	281	300	150	75
b															
c															

a = native starch in percent by weight

b = 500 wt. % water in relation to fiber material (dry substance)

c = 100% fiber material

Fig. 4b

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	$x_1$	$x_2$	$x_3$	$x_4$	$x_5$	$x_6$	$x_7$	$x_8$	$x_9$	$x_{10}$	$x_{11}$	$x_{12}$	$x_{13}$	$x_{14}$	$x_{15}$
a	18,7	28,2	37,3	47	56,4	65,8	75	84,3	93,3	102,9	112,3	122	131,4	140,8	150
d	6,3	9,4	12,7	15,7	18,8	21,9	25	28,3	31,3	34,9	37,7	40,7	41,8	46,9	50
e	24,9	37,8	50	62,7	75,2	87,7	100	112,7	124,7	137,8	150	162,7	175,2	187,7	200

Fig. 5a

 $x_{1-15}$  test sample

a in wt. % native starch

d in wt. % pregelatinized starch

e in wt. % total starch

b = 250 wt. % water (in relation to dry mass  
of fiber material)

c = 100 wt. % fiber material

## ratios

test sample	$y_1$	$y_2$	$y_3$	$y_4$	$y_5$	$y_6$	$y_7$	$y_8$	$y_9$	$y_{10}$	$y_{11}$	$y_{12}$	$y_{13}$	$y_{14}$	$y_{15}$
B	94	113	131	150	169	188	206	225	244	283	281	300	319	350	375
J	75	31	38	44	50	58	63	68	75	81	88	94	100	100	100
E	50	75	100	125	150	175	200	225	250	275	300	325	350	375	400

a = native starch in wt. %  
 d = pregelatinized starch in wt. %  
 e = total starch in wt. %  
 b = 500 wt. % water in relation to fiber material (dry substance)  
 c = 100% fiber material

Fig. 5b

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Grade	Long fibers/ fiber bundles [mm]
1	0.96 - 1.44
2	1.92 - 2.40
3	2.40 - 2.88
4	0.72 - 2.16
5	3.06 - 3.57
6	2.55 - 4.59
7	0.24 - 1.68
8	0.24 - 4.32

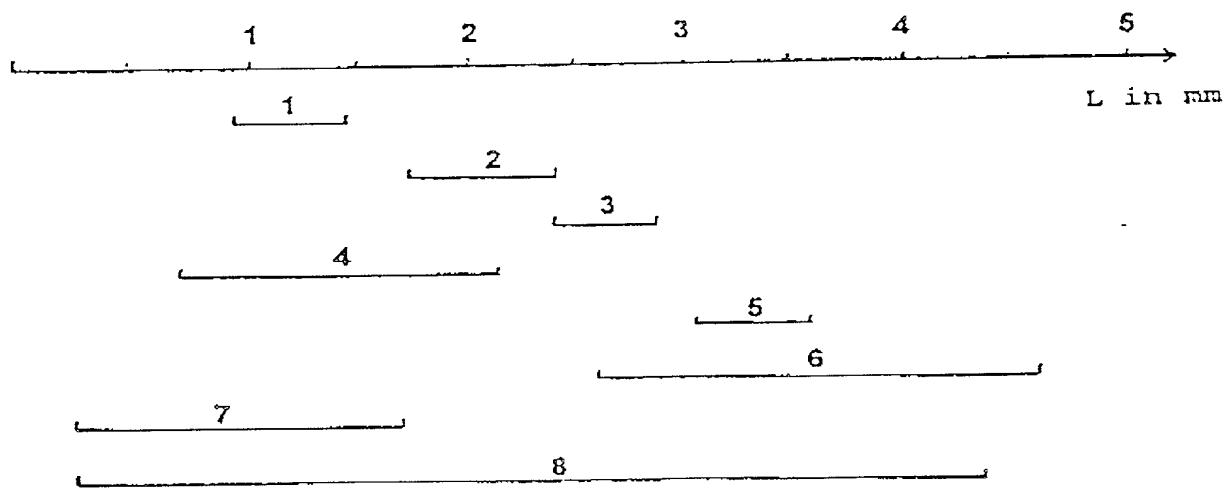


Fig. 6

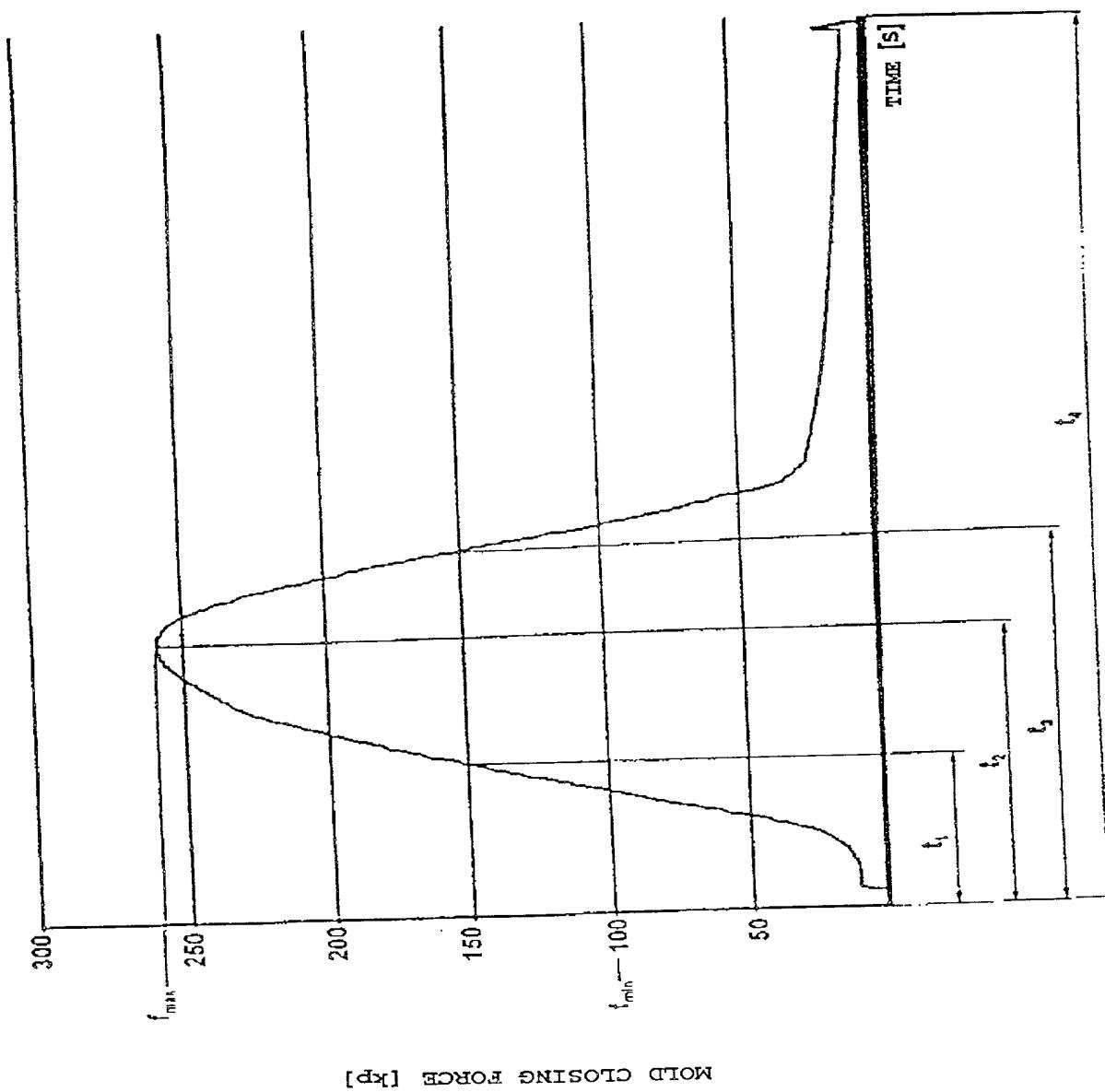
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Fig. 7



Use of fibers (fiber bundles) graded by fiber length according to Fig. 6

mold depth	~ 30 mm								~ 50 mm							
	1	2	3	4	5	6	7	8	1	2	3	4	5	6	7	8
fiber length	+	+	+	+	-	+	-	+	+	+	+	+	+	+	+	-
surface/ texture	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
strength/ stability	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
elasticity/ structure	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

mold depth	~ 80 mm								> 80 mm							
	1	2	3	4	5	6	7	8	1	2	3	4	5	6	7	8
fiber length	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
surface/ texture	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
strength/ stability	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
elasticity/ structure	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

+ molded body according to requirements

- molded body not according to requirements

Fig. 8

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Use of fiber mixtures of different fiber length according to Fig. 6

mold depth	- 30 mm			- 50 mm			- 80 mm			> 80 mm		
	7 + 4	4 + 2	7 + 2 + 3	4 + 2 + 3	8	7 + 2 + 3 + 5	8	7 + 2 + 6	8	7 + 2 + 6	8	7 + 2 + 6
combination of fiber lengths according to Fig. 6	-	+	-	+	+	-	+	-	+	-	+	-
surface/texture	+	+	+	+	+	+	+	+	+	+	+	+
strength/stability	-	+	+	+	+	-	+	+	+	+	+	+
elasticity/structure	-	+	+	+	+	-	+	+	+	+	+	+
fiber material/starch	60 : 40		55 : 45			50 : 50		45 : 55				
starch/water	0,4 : 1		0,4 : 1			0,4 : 1		0,3 : 1				

+ molded body according to requirements

- molded body not according to requirements

Fig. 9

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in wt. %	$x_1$	$x_2$	$x_3$	$x_4$	$x_5$	$x_6$	$x_7$	$x_8$	$x_9$	$x_{10}$	$x_{11}$	$x_{12}$	$x_{13}$	$x_{14}$	$x_{15}$
fiber material to total mass	26,7	25,8	25	24,2	23,5	22,8	22,2	21,6	21,1	20,5	20	19,5	19	10,6	18,2
total starch to total mass	6,6	9,7	12,5	15,2	17,7	20,1	22,2	24,3	26,2	28,2	10	11,7	13,4	34,9	16,1
water to total mass	66,7	64,5	62,5	60,6	59,8	57,1	55,6	54,1	52,7	51,3	50	48,8	47,6	46,5	45,5
pregelatinized starch to total mass	1,6	2,4	1,2	1,8	4,4	5	5,5	6,1	6,6	7,1	7,5	7,9	8,4	8,7	9

 $x_{15}$  test sample

Fig. 10

## Percent by weight in mass

	$\gamma_1$	$\gamma_2$	$\gamma_3$	$\gamma_4$	$\gamma_5$	$\gamma_6$	$\gamma_7$	$\gamma_8$	$\gamma_9$	$\gamma_{10}$	$\gamma_{11}$	$\gamma_{12}$	$\gamma_{13}$	$\gamma_{14}$	$\gamma_{15}$	
in wt. %	14.3	13.8	11.3	12.9	12.5	12.1	11.0	11.4	11.1	10.8	10.5	10.3	10.0	11.8	12.9	
fiber/mass	14.3	17.2	17.2	17.2	22.6	25.0	27.2	29.1	31.4	33.3	35.1	36.0	36.5	40.0	28.4	22.6
total starch/mass	10.7	4.3	5.0	5.0	6.3	6.8	7.4	7.9	8.3	8.9	9.2	9.6	10.0	11.4	12.9	
pregel. starch/mass	71.4	69.0	66.7	64.5	62.5	60.8	58.8	57.1	55.8	54.1	52.0	51.3	50.0	58.8	61.5	
water/mass																

 $\gamma_{15}$  = test sample

Fig. 11

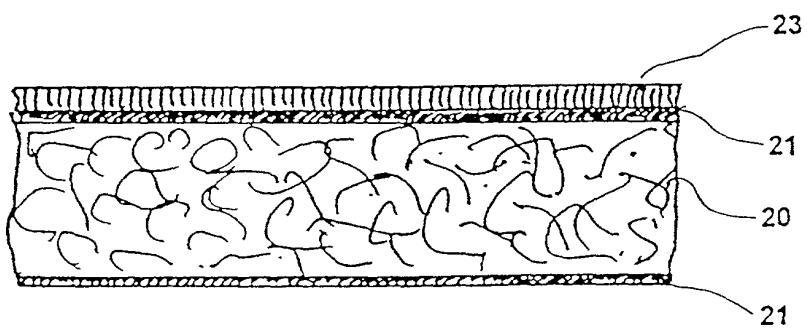


Fig. 12

Trays      dimensions      112 x 200 x 17.5 mm

Pots      dimensions      Ø 125 mm, vol. 500 ml, height 76 mm

Recipe:    Y14

Coating:    cellulose acetate (CA)

TS: 4.5% - 15 wt. % dry substance in spray solution

$\eta$ : 20 - 4000 mPas (viscosity)

Application: spraying, casting, dipping

Layers:    1 - 3 (quantity)

Solvent:    acetone

Shape	Thick- ness	Coating	Method	Resistance		
				water 100°C	oil (cold)	water (cold)
				1h	3 days	3 days
pot	89 $\mu$ m	3.8 g	casting	+	+	+
tray	79 $\mu$ m	2.3 g	casting	+	+	+
pot	65 $\mu$ m	2.8 g	spraying	+	+	+
tray	68 $\mu$ m	2.0 g	spraying	+	+	+
tray	58 $\mu$ m	1.7 g	spraying	+	+	+
pot	34 $\mu$ m	1.5 g	spraying	-	-	-
tray	27 $\mu$ m	0.8 g	spraying	-	-	-

Fig. 13

Trays dimensions 112 x 200 x 17.5 mm

Pots dimensions Ø 125 mm, vol. 500 ml, height 76 mm

Recipe: Y14

Coating: cellulose acetate propionate(CAP)

TS: 9% - 20 wt. % dry substance in spray solution

$\eta$ : 200 - 6000 mPas (viscosity)

Application: spraying, casting, dipping

Layers: 1 - 3 (quantity)

Solvent: acetone

Shape	Thickness	Method	Resistance			
			water 100°C 1h	oil cold 3 days	water cold 3 days	water cold 3 days
pot	88 $\mu$ m	casting	+	+	+	+
tray	88 $\mu$ m	casting	+	+	+	+
pot	58 $\mu$ m	spraying	+	+	+	+
tray	70 $\mu$ m	spraying	+	+	+	+
tray	56 $\mu$ m	spraying	+	+	+	+
pot	33 $\mu$ m	spraying	-	-	-	-
tray	22 $\mu$ m	spraying	-	-	-	-

Fig. 14

Trays dimensions 112 x 200 x 17.5 mm

Pots dimensions Ø 125 mm, vol. 500 ml, height 76 mm

No.	Foil	Thick-ness	Deep-drawing quality in tray	Deep-drawing quality in tray	Resistance		
					water 100°C	oil cold	water cold
1	poly-ester amide	100 µm	+	-	-	+	+
		150 µm	+	-	—	+	+
2	poly-ester	70 µm	+	-	—	+	+
3	poly-lactic acid (rigid)	50 µm	-	-	-	+	+
		100 µm	-	-	-	+	+
4	poly-lactic acid (elast.)	50 µm	+	-	+	+	+
		100 µm	+	+	+	+	+

Foil Melting point

1 approx. 120°C

2 approx. 85°C

3 approx. 115°C

4 approx. 130°C

## Cellulose acetate / Cellulose acetate propionate

Softener 10-30 wt.%					without softener
Diethyl-phthalate		Triacetin	Tributyl citrate	Acetyl tributyl citrate	
CA	V+/H+	V+/H+	V-/H-	V-/H-	H O
CAP	V+/H+	V+/H+	V+/H+	V+/H+	H +

Key: + = good O = medium - = poor  
 V = compatibility H = adhesion

Fig. 16

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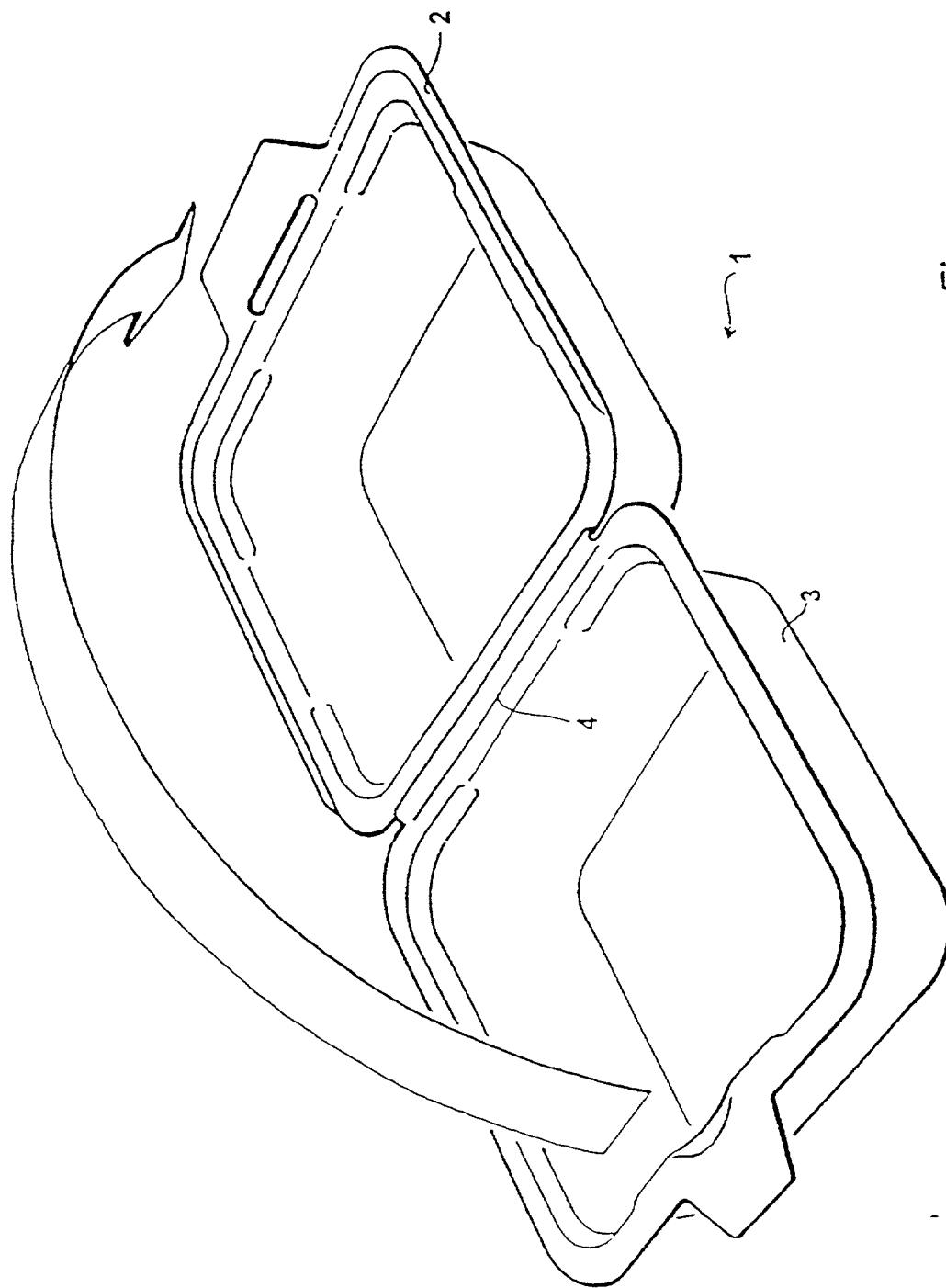


Fig. 17

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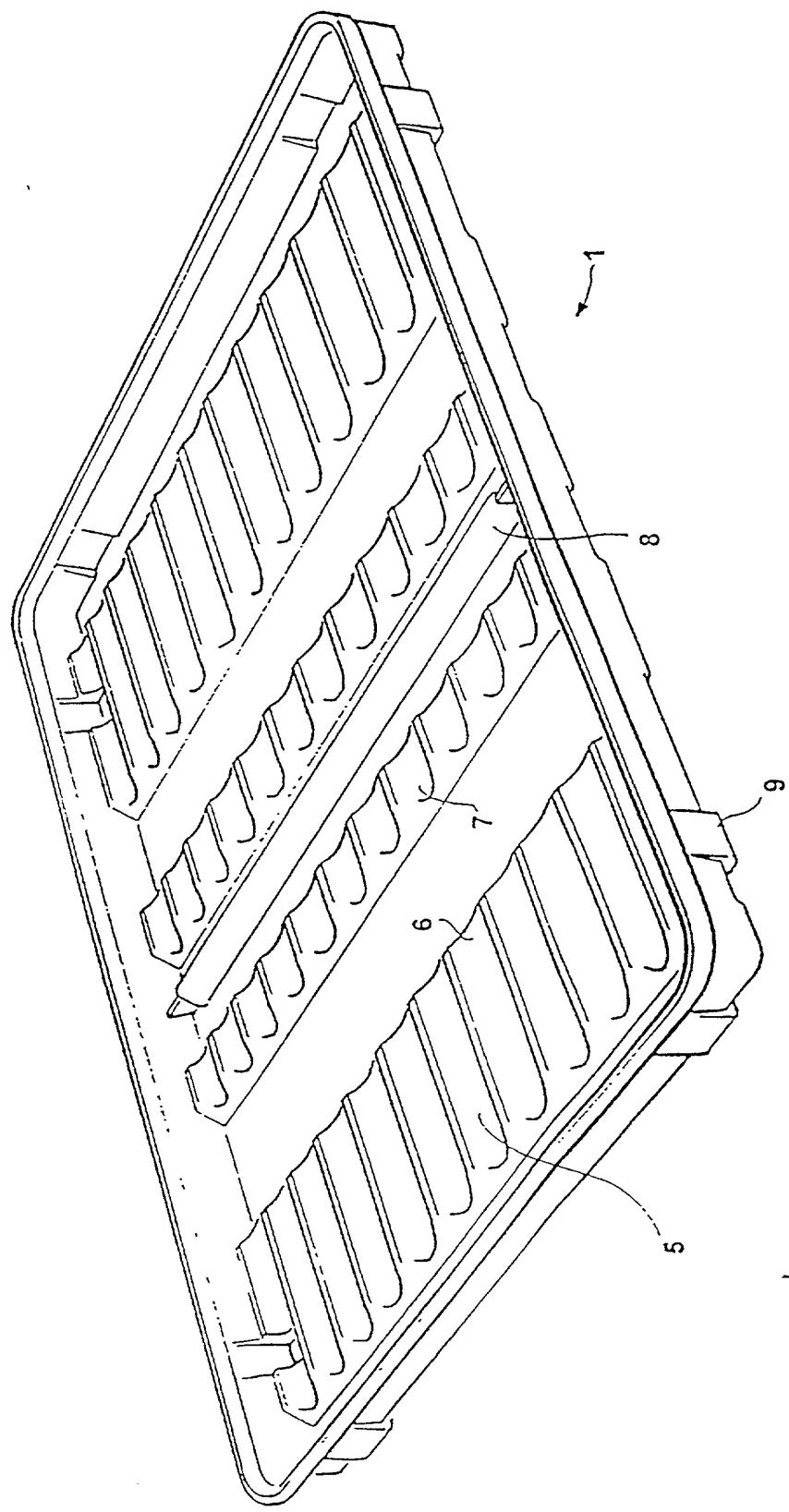


Fig. 18

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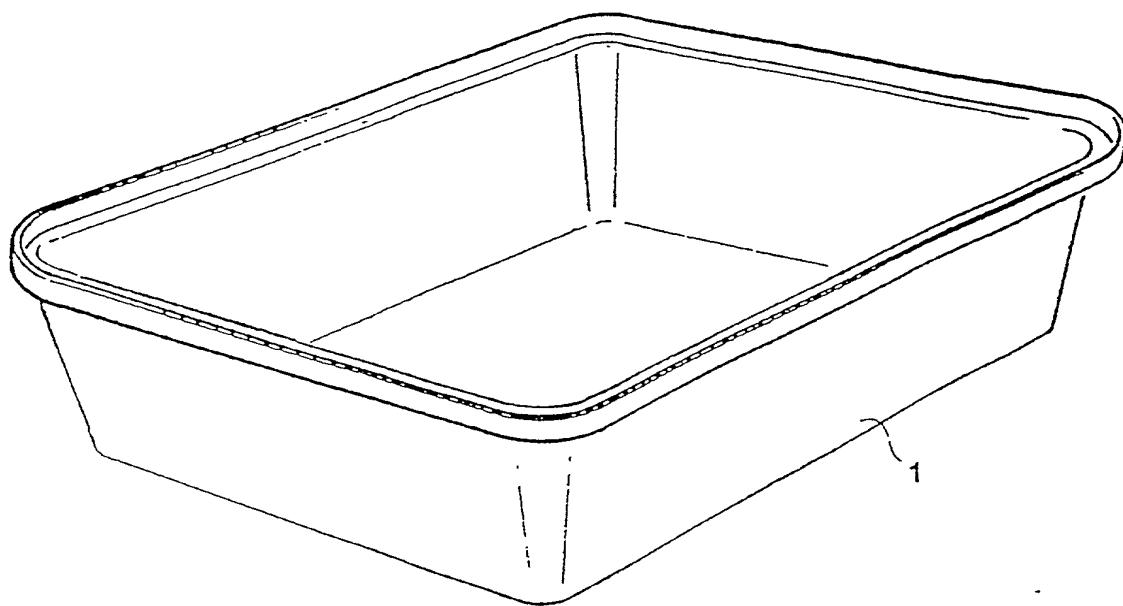


Fig. 19

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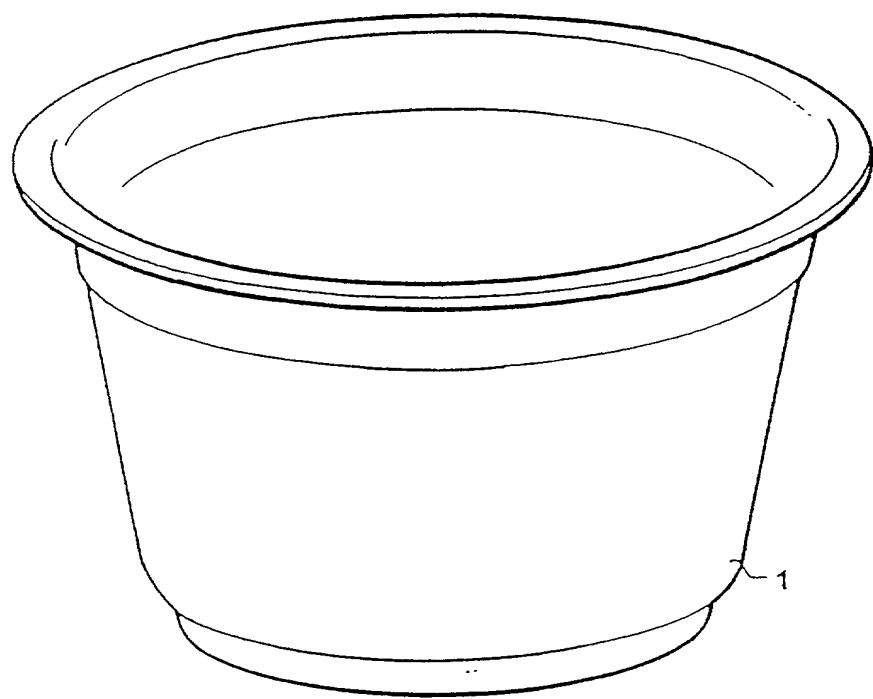


Fig. 20

UNITED STATES OF AMERICA  
COMBINED DECLARATION AND POWER OF ATTORNEY  
FOR PATENT APPLICATION

FILE NO. GK-GEY-1020

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I verily believe that I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural inventors are named) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

PROCESS FOR PRODUCING MOULDINGS WITH A BARRIER LAYER MADE OF BIODEGRADABLE MATERIAL  
AND MOULDINGS PRODUCED ACCORDING TO THIS PROCESS

the specification of which

is attached hereto.

was filed on \_\_\_\_\_ as United States patent application Serial Number \_\_\_\_\_

was filed on September 12, 1996 as PCT international patent application No. PCT/EP96/04016 and was amended on \_\_\_\_\_ (if any).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose all information known to be material to patentability in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

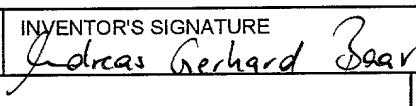
COUNTRY	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 35 U.S.C. 119
WO	PCT/EP95/03592	12 September 1995	YES <input checked="" type="checkbox"/> NO _____
			YES _____ NO _____

I hereby appoint McAULAY FISHER NISSEN GOLDBERG & KIEL and the members of the firm: Lloyd McAulay, Reg. No. 20,423; J. Harold Nissen, Reg. No. 17,283; Jules E. Goldberg, Reg. No. 24,408; Gerald H. Kiel, Reg. No. 25,116; William J. Sapone, Reg. No. 32,518; Francis C. Hand, Reg. No. 22,280; Eugene LeDonne, Reg. No. 35,930; and Mark Montague, Reg. No. 36,612; as attorneys with full power of substitution and revocation to prosecute all business in the Patent & Trademark Office connected therewith and to receive all correspondence.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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COMBINED DECLARATION AND POWER OF ATTORNEY  
FOR PATENT APPLICATION (continued )

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RESIDENCE		COUNTRY OF CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SEVENTH JOINT INVENTOR (IF ANY)		INVENTOR'S SIGNATURE	DATE
RESIDENCE		COUNTRY OF CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF EIGHTH JOINT INVENTOR (IF ANY)		INVENTOR'S SIGNATURE	DATE
RESIDENCE		COUNTRY OF CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF NINTH JOINT INVENTOR (IF ANY)		INVENTOR'S SIGNATURE	DATE
RESIDENCE		COUNTRY OF CITIZENSHIP	
POST OFFICE ADDRESS			